

Physics of plasma jets and interaction with surfaces: modelling and experiments

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Online Low Temperature Plasma (OLTP) seminar, July 12th 2022

Co-workers on plasma jet research

Laboratoire de Physique des Plasmas (LPP), École Polytechnique, France:

- Anne Bourdon.
- Olivier Guaitella.
- Elmar Slikboer.

Elementary Processes in Gas Discharges (EPG),
Eindhoven University of Technology (TU/e), The Netherlands:

- Ana Sobota.
- Marlous Hofmans.

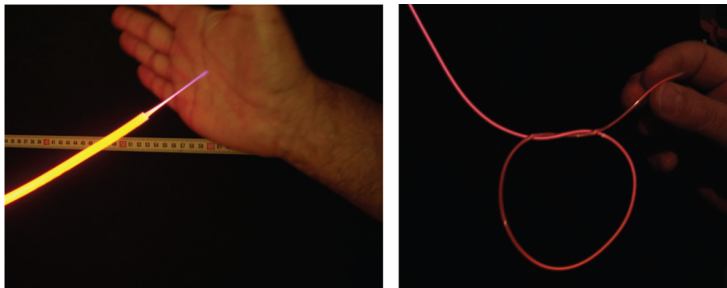
Centre for Plasma Microbiology, The University of Liverpool, United Kingdom:

- Elmar Slikboer.

Department of Physical Electronics, Masaryk University, Czech Republic:

- Zdeněk Bonaventura.

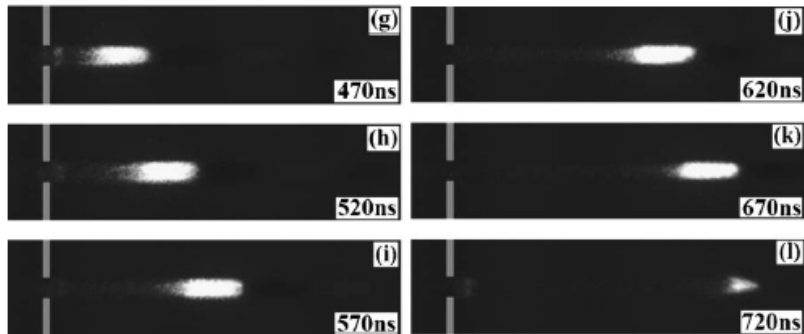
Physics of plasma jets and interaction with surfaces: **review** on modelling and experiments, *Plasma Sources Sci. Technol.* 31 (2022) 053001



E Robert *et al.*, *Plasma Process. Polym.* 6 (2009) 795

- Atmospheric pressure room-temperature discharges.
- Discharge guided by noble gas flow (e.g. He), ignites in dielectric tube and propagates in air towards a target.
- Applied on surfaces and biological tissues.

Plasma jets - ionization wave propagation



M Teschke *et al.*, *IEEE Trans. Plasma Sci.* 33 (2005) 310

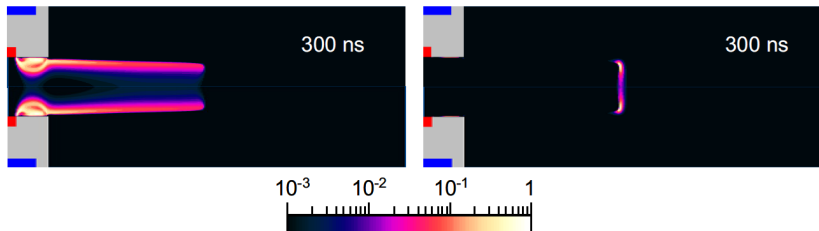
X Lu and M Laroussi, *J. Appl. Phys.* 100 (2006) 063302

- At naked eye, continuous plasma plume luminosity.
- First observations in 2005: Jet luminosity consists of repetitive discrete luminous fronts travelling at $\sim 10\text{-}100 \text{ km.s}^{-1}$ with radius $\sim 1 \text{ mm}$.

Plasma jets - ionization wave propagation

n_e = electron density

S_e = electron-impact ionization source term



G Naidis, *J. Phys. D: Appl. Phys.* 43 (2010) 402001

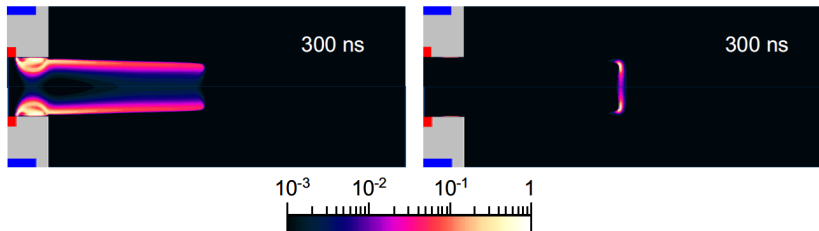
J-P Boeuf *et al.*, *J. Phys. D: Appl. Phys.* 46 (2013) 015201

- Simulations of jet propagation in the noble gas channel without air mixing.
- Propagation as streamer-like discharges, driven by their own space-charge electric field.

Plasma jets - ionization wave propagation

n_e = electron density

S_e = electron-impact ionization source term



G Naidis, *J. Phys. D: Appl. Phys.* 43 (2010) 402001

J-P Boeuf *et al.*, *J. Phys. D: Appl. Phys.* 46 (2013) 015201

- Ionization front with localized charge production and plasma channel behind it.
- **Repeatable stable IW propagation:**
Ideal testbed to study transient discharge dynamics and surface interaction.

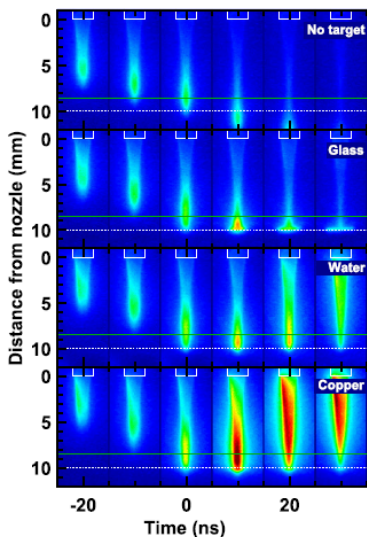
Plasma jets and interaction with targets



T Darny *et al.*, 6th *International Conference on Plasma Medicine* (2016)

- Treated surfaces with different electrical character:
 - Conductive / dielectric, ϵ_r .
 - Grounded / floating potential.
- Jets affect targets: reactive species, radiation, electric field and charges.
- Targets affect discharge dynamics depending on electrical character.

Plasma jets and interaction with targets



- **Physics of discharges** is affected by electrical character of **target**.

- Dielectric of different permittivities.
- Metallic.
- Grounded or floating.

S Norberg *et al.*, *J. Appl. Phys.* 118 (2015) 013301

Y Yue *et al.*, *Plasma Sources Sci. Technol.* 27 (2018) 064001

L Ji *et al.*, *J. Appl. Phys.* 123 (2018) 183302

B Klarenaar *et al.*, *Plasma Sources Sci. Technol.* 27 (2018) 085004

Review on physics of plasma jets and interaction with surfaces:

- Physical characterization of plasma jets
 - Numerical modelling
 - Diagnostics and experimental benchmarking
- Insights from comparisons between simulations and experiments
 - Jet interacting with dielectric target
 - Jet interacting with metallic targets
- Outlook on future of physics of plasma jets and interaction with surfaces

Plasma jets and interaction with targets

Physical characterization by:

- **Experimental diagnostics.**
- **Numerical simulations.**
- Qualitative comparisons between experiments and simulations: discharge structure.
W Ning et al., J. Phys. D: Appl. Phys. 51 (2018) 125204
C Lazarou et al., Plasma Sources Sci. Technol. 27 (2018) 105007
- Quantitative comparisons of macroscopic parameters: velocity of discharge propagation.
L Ji et al., J. Appl. Phys. 123 (2018) 183302
- **Recently, quantitative comparisons, temporally and spatially resolved, between experiments and simulations.**

Modelling plasma jets

0D and 1D models to focus on chemical kinetics in plasma jets.

W V Gaens and A Bogaerts, *J. Phys. D: Appl. Phys.* 46 (2013) 275201

Y He *et al.*, *Eur. Phys. J. D* 75 (2021) 2021

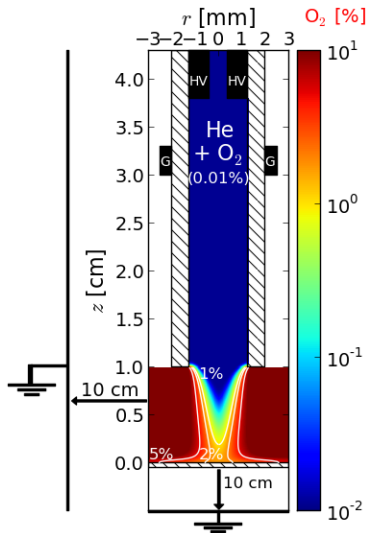
For filamentary transient discharge physics, usually **2D fluid models**, based on streamer discharges.

G V Naidis, *J. Phys. D: Appl. Phys.* 43 (2010) 402001

D Breden *et al.*, *Plasma Sources Sci. Technol.* 21 (2012) 034011

- Setup: tube, electrodes, targets, grounded surfaces.
- Noble gas flow into air/shielding gases: spatial variations of gas composition.

N Y Babaeva and G V Naidis, *J. Phys. D: Appl. Phys.* 54 (2021) 223002



P Viegas and A Bourdon, *Plasma Chem. Plasma Process.* 40 (2020) 661-683

Modelling plasma jets: plasma-flow coupling

2D fluid models with noble gas flow and heterogeneous gas composition in the plume:

- No mixing assumption: uniform noble gas channel surrounded by air.

G V Naidis, *J. Phys. D: Appl. Phys.* 43 (2010) 402001

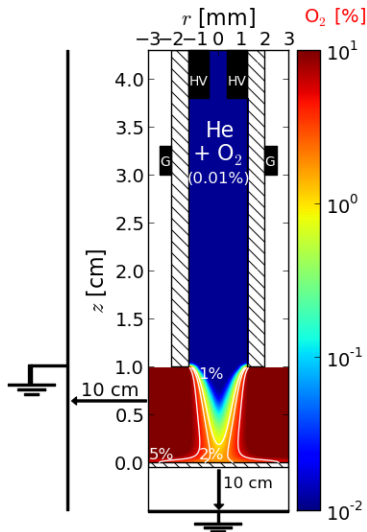
J-P Boeuf *et al.*, *J. Phys. D: Appl. Phys.* 46 (2013) 015201

- Two-way plasma-flow coupling: Effects of the plasma on the flow through electrohydrodynamic force and heating.

P K Papadopoulos *et al.*, *J. Phys. D: Appl. Phys.* 47 (2014) 425203

A M Lietz *et al.*, *Appl. Phys. Lett.* 111 (2017) 114101

- Most common **static flow approximation**.

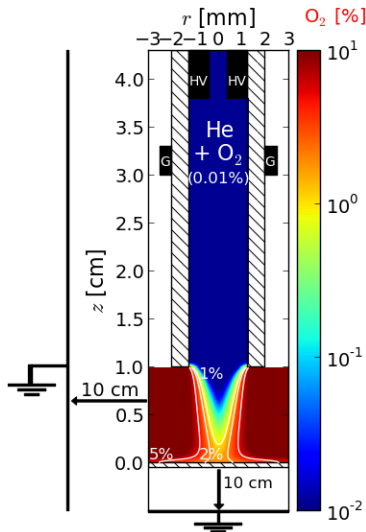


P Viegas and A Bourdon, *Plasma Chem. Plasma Process.* **40** (2020) 661-683

Modelling plasma jets: coupling with pre-calculated flow

Static flow models:

- Helium with impurities, flowing into air or shielding gases (N_2 , O_2 , H_2O) at ~ 1 L/min.
- Flow velocity of a few $\mu\text{m}/\mu\text{s}$. Static approximation in plasma μs timescale.
- Flow calculations (Navier-Stokes solution) set densities of, e.g., **He**, N_2 and O_2 .
- Impact on chemistry, electron rate coefficients, transport parameters and photoionization.
- Assumption: no effect of plasma on flow. Low heating in kHz pulses in He.



P Viegas and A Bourdon, *Plasma Chem. Plasma Process.* **40** (2020) 661-683

2D fluid models for discharge dynamics in plasma jets:

- Simplest: Drift-diffusion models with local electric field approximation (LFA).
- More accurate models use local mean electron energy approximation (LMEA).
Continuity equations are solved for charged species and electron mean energy:

$$\frac{\partial n_k}{\partial t} + \nabla \cdot \mathbf{j}_k = S_k \quad (1)$$

$$\frac{\partial}{\partial t}(n_e \epsilon_m) + \nabla \cdot \mathbf{j}_e = -|q_e| \mathbf{E} \cdot \mathbf{j}_e - \Theta_e \quad (2)$$

Drift-diffusion approximation:

$$\mathbf{j}_k = \mu_k n_k \mathbf{E} - D_k \nabla n_k \quad (3)$$

- Chemical kinetics for, e.g., He-N₂-O₂ mixtures.
- Electron kinetics coefficients often precalculated and tabulated for several He-N₂-O₂ mixtures and ϵ_m values with EBE solver.
Interpolated/fitted as $f(\text{mix}, \epsilon_m)$ in each cell and at each timestep.

- **Poisson's equation** to calculate EF distribution:

$$\epsilon_0 \nabla \cdot (\epsilon_r \nabla V) = -\rho - \sigma \delta_s \quad (4)$$

$$\mathbf{E} = -\nabla V; \rho = \sum_k q_k n_k \quad (5)$$

- **Surface charge density** σ often accumulated on dielectric surfaces (tube and target):
 - Charge flux via electric drift and thermal motion.
 - Surface reactions of charges: electron absorption, ion neutralization, secondary electron emission (SEE) via ion and photon impact ($\gamma_{pe} = 0.01$, $\gamma_{se} = 0.1$).
- Single discharge simulation in **repetitive conditions**:
Memory effects in volume and surfaces.
- Initial conditions:
Typical assumption of neutralized surfaces between each pulse.

- Streamer-like propagation requires free charges ahead of discharge front: **seed charges**.
- Single discharge simulation in **repetitive conditions**:
Memory effects in volume and surfaces.
- Initial conditions:
Typically $\sim 10^9 \text{ cm}^{-3}$ preionization (e, N_2^+ , O_2^+) for repetitive kHz conditions.
G V Naidis, *J. Phys. D: Appl. Phys.* 44 (2011) 215203
X Lu and K Ostrikov, *Appl. Phys. Rev.* 5 (2018) 031102
- Photoionization: at p_{atm} , ionizing radiation mostly from He_2^* .
G V Naidis, *J. Phys. D: Appl. Phys.* 43 (2010) 402001
A M Lietz and M J Kushner, *Plasma Sources Sci. Technol.* **27** (2018) 105020

Electron density n_e and electric field magnitude E_t during a rectangular pulse, from $t = 0$ to $t = 800$ ns

- Ignition at inner electrode.
- Discharge propagation in tube.
- Discharge propagation in plume close to axis (region with air fraction below 5%).
- Impact and spreading on a dielectric target with $\epsilon_r = 56$.

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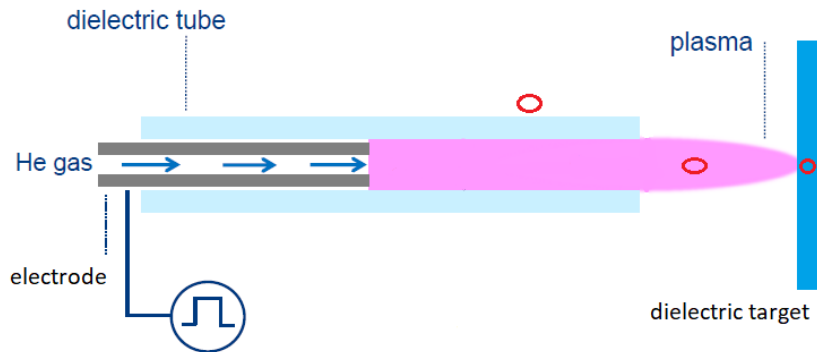
First, measurements of discharge current and light emission intensity.

Recently, measurements of **electric field (EF) and electron density**:

- **Key quantities** for discharge dynamics (ionization, drift of charged particles, charge deposition on surfaces).
- These quantities are affected by jet source parameters: flow, geometry, excitation source.
- Drive the production of active species of interest for plasma jet applications.
- Opportunity to carry out spatially and temporally resolved quantitative **comparisons** between simulations and experiments.

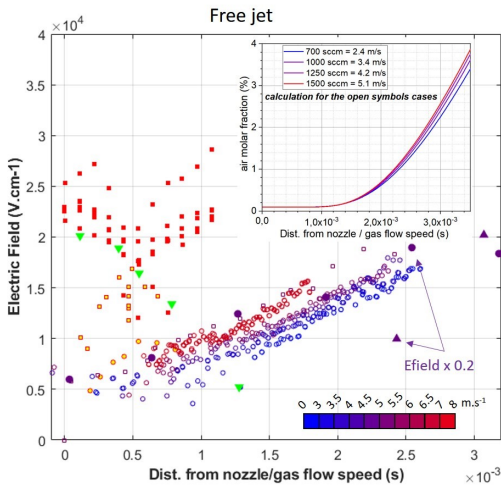
EF diagnostics in plasma jets

Different spatially and temporally resolved experimental techniques assessing EF in different locations



Benchmarking **peak EF at the discharge front as $f(\text{mix})$** in freely expanding plasma plume.

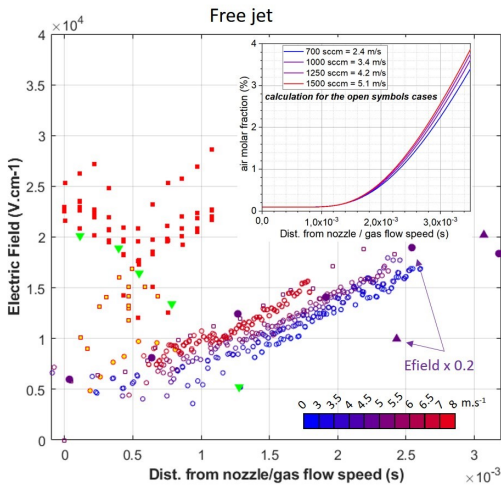
- OES on Stark-shifted forbidden He lines
A Sobota *et al.*, *Plasma Sources Sci. Technol.* 25 (2016) 065026
- Electric Field Induced Second Harmonic Generation (E-FISH) in Ar
B M Goldberg *et al.*, *Opt. Lett.* 44 (2019) 3853
- Line intensity ratio from OES on N_2
A Begum *et al.*, *AIP Adv.* 3 (2013) 062117



P Viegas *et al.*, *PSST* 31 (2022) 053001

Benchmarking **peak EF at the discharge front as $f(\text{mix})$** in freely expanding plasma plume.

- Different experiments/sources in He jets show similar increase of peak EF with air molar fraction in plume.
- Same trend but 5 times higher EF by N_2 line ratio (●).
- Opposite trend in Ar jet by E-FISH (▼).
- Different values/trend with biased cylinder around the jet (■).

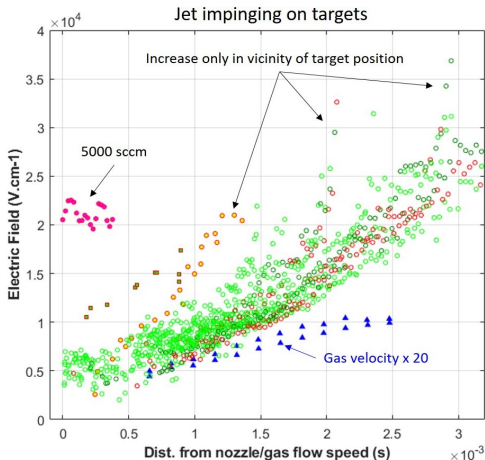


P Viegas *et al.*, *PSST* 31 (2022) 053001

EF diagnostics in plasma jets

Benchmarking **peak EF at the discharge front as $f(\text{mix})$** in jets with targets.

- OES on Stark-shifted forbidden He lines
G Sretenovic *et al.*, *Appl. Phys. Lett.* 99 (2011) 161502
G Sretenovic *et al.*, *J. Phys. D: Appl. Phys.* 47 (2014) 102001
A Sobota *et al.*, *Plasma Sources Sci. Technol.* 28 (2019) 045003
M Mirzaee *et al.*, *Phys. Plasmas* 27 (2020) 123505
- Electric Field Induced Second Harmonic Generation (E-FISH) in He
K Orr *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 035019



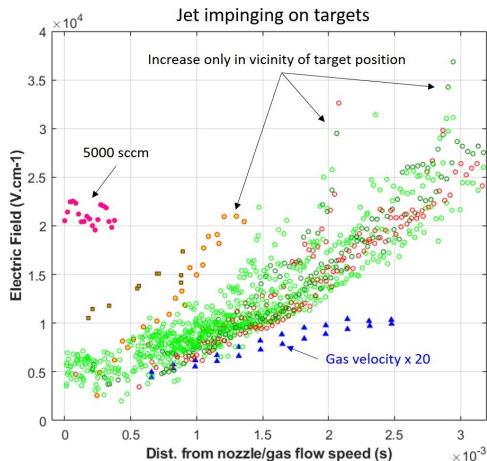
P Viegas *et al.*, *PSST* 31 (2022) 053001

EF diagnostics in plasma jets

Benchmarking **peak EF at the discharge front as $f(\text{mix})$** in jets with targets.

- Same increase with air fraction as in He free jet.
- Sharp increase of EF close to different targets (dielectric and metallic)

In general, increase of confidence in EF values and trends by **benchmarking different measurements** of peak EF at the discharge front in different jets with and without targets.



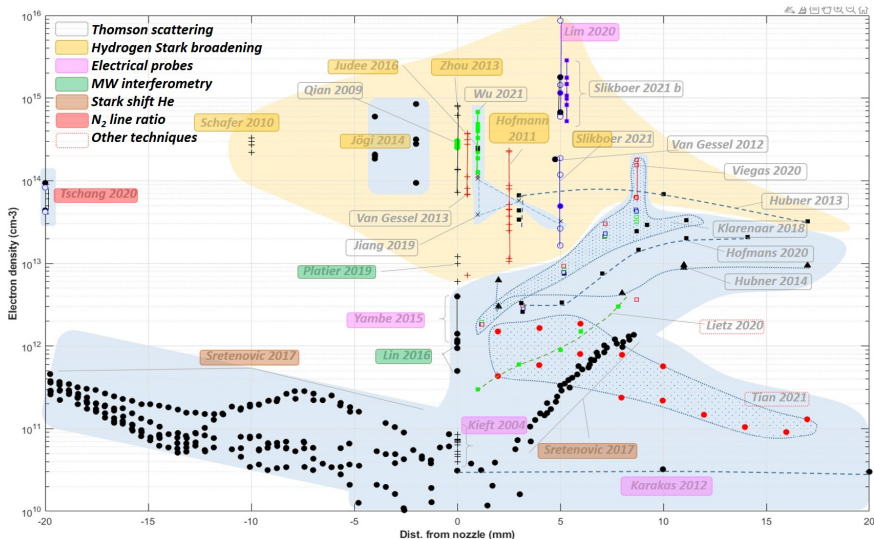
P Viegas *et al.*, *PSST* 31 (2022) 053001

Scarce data on electron temperature.

For **electron density** (n_e):

- Difficult measurements: transient nature, small size and jitter of jets.
- More methods than for the EF: from current measurements to spectroscopic and laser-based diagnostics.
- Measured n_e for more sources and gases than the EF: high variability of n_e .
- Data spanning over 6 orders of magnitude.

Electron density diagnostics in plasma jets

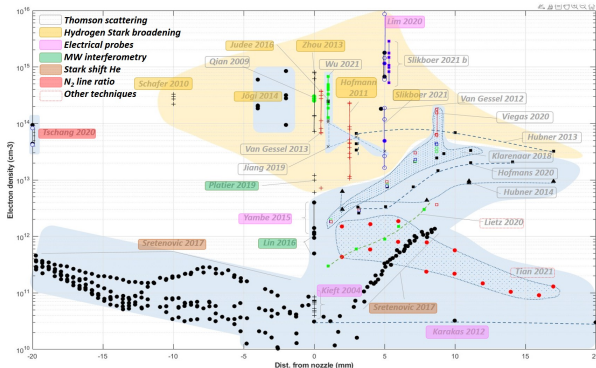


P Viegas et al., *Plasma Sources Sci. Technol.* 31 (2022) 053001

Electron density diagnostics in plasma jets

Benchmarking **maximum $n_e(z)$ in the plasma** in jets with and without targets.

- n_e in Ar jets on average higher than in He jets.
- In He, n_e profile follows peak EF profile.
- RF jets generate lower n_e than kHz AC and pulsed.
- Using grounded targets yields higher densities than floating targets.



More difficult to benchmark n_e than EF.

P Viegas et al., *Plasma Sources Sci. Technol.* 31 (2022) 053001

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Characterization of plasma jets by comparisons

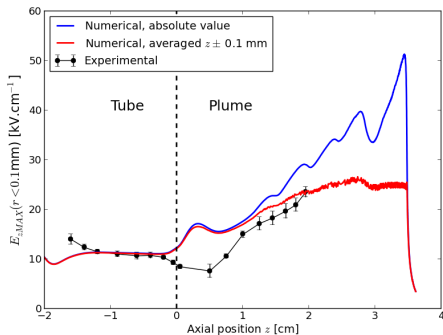
Quantitative spatially and temporally-resolved comparisons between experiments and simulations:

- Understand discharge dynamics.
- Validate models and different experimental techniques to increase confidence.
- Simulations can be used in a complementary way to evaluate parameters where it is difficult to measure.
- Identify differences in the quantities under comparison.

Recently, few works comparing numerical and experimental results for the same jet setup.

Comparisons in free jet (He kHz pulsed): summary

- Qualitative agreement on discharge propagation and structure.
 - Excellent agreement on discharge propagation velocity in the tube and in the plume.
 - **Agreement of simulations with measurements on peak EF:** around $10 \text{ kV}\cdot\text{cm}^{-1}$ in tube and increasing in plume.
- N Y Babaeva and G V Naidis, *J. Phys. D: Appl. Phys.* 54 (2021) 223002
- High EF ($\sim 20 \text{ kV}\cdot\text{cm}^{-1}$) in the discharge front due to charge separation.
 - Importance of taking into account **spatial and temporal averages** when comparing simulations with measurements.

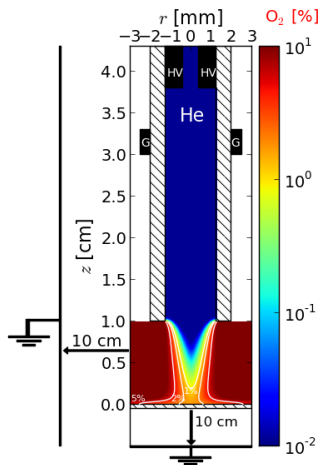


M Hofmans *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 034003

EF measurements inside a dielectric BSO target

Laboratoire de Physique des Plasmas, E Slikboer, *PhD thesis* (2018)

- Jet generated by rectangular positive pulse of variable width (for example $1 \mu\text{s}$) applied to electrode inside the tube.
- Flow of 1.0 L/min of Helium through the pyrex tube into air atmosphere.
- Interaction between plasma plume and dielectric BSO crystal ($\epsilon_r = 56$) at 1 cm from the end of the tube.

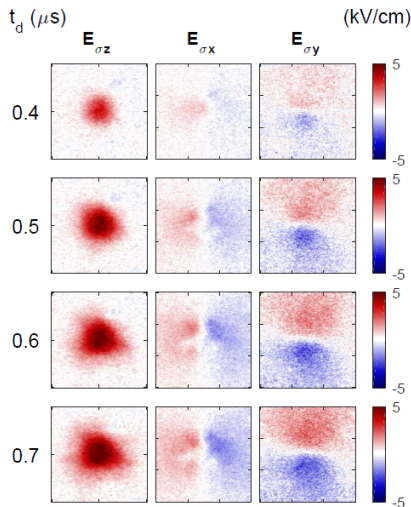


E Slikboer *et al.*, *Plasma Sources Sci. Technol.* 28 (2019) 095016

EF measurements inside a dielectric BSO target

Laboratoire de Physique des Plasmas, E Slikboer, *PhD thesis* (2018)

- During plasma exposure, charges deposited on the target's surface.
- **EF inside the target** measured through Mueller polarimetry.
- Spatial profiles of EF components **averaged inside the target** through 0.5 mm thickness.
- EF inside target $\sim 4 \text{ kV}\cdot\text{cm}^{-1} < \sim 20 \text{ kV}\cdot\text{cm}^{-1}$ in the discharge front.
Why?



P Viegas *et al.*, *Plasma Sources Sci. Technol.* 27 (2018) 094002

Simulation of plasma jet interaction with dielectric target

Discharge propagation and spreading ($V_P = +6$ kV and $t_f = 1.1$ μ s)

- End of tube at $t = 210$ ns.
- Centered structure of propagation, as in experiments.
- Centered impact on the target surface at $t = 290$ ns.
- Spreading on target surface until end of pulse at $t = 1100$ ns.

Simulation of plasma jet interaction with dielectric target

Discharge propagation and spreading ($V_P = +6$ kV and $t_f = 1.1$ μ S)

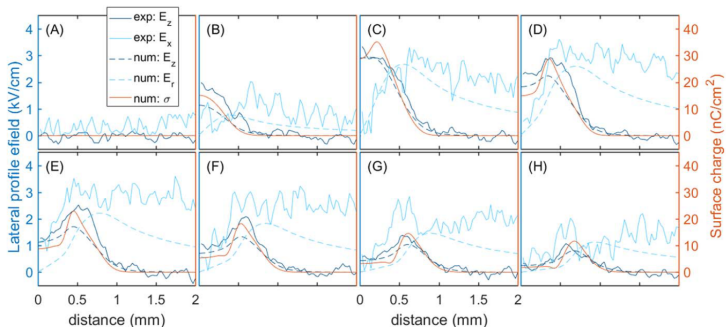
E_t = magnitude of the EF

Spreading of the discharge on the target and end of pulse:

- EF partially penetrates inside the target.
- Spreading slows down and E_t decreases after end of pulse at $t = 1100$ ns.

Electric field comparisons in dielectric BSO target

$V_P = +6$ kV and $t_f = 320$ ns



E Slikboer *et al.*, *Plasma Sources Sci. Technol.* 28 (2019) 095016

EF components inside dielectric target of $\epsilon_r = 56$ after discharge impact, for short pulse. Radial profiles at different times of E_z and E_r , averaged over the target thickness, and of surface charge density σ .

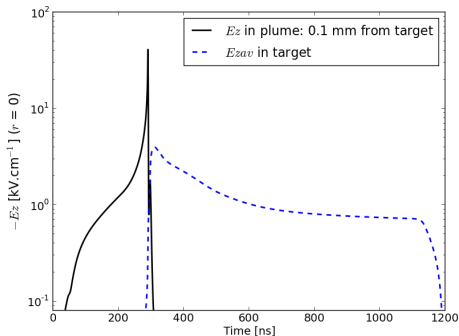
- First temporally and spatially-resolved quantitative comparisons between simulations and experiments, with excellent agreement.
- Lower EF than in plume confirmed by simulations.

Electric field comparisons in dielectric BSO target

$V_P = +6$ kV, $t_f = 1.1$ μ s

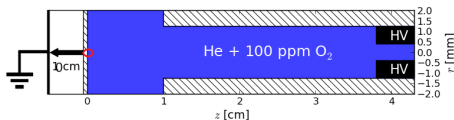
Why is the EF in the target lower than the EF in the plume?

- Peak EF in the plume, due to charge separation in volume, in short timescale.
- EF inside the target in longer timescale but one order of magnitude **lower than EF in the plume** impinging the target, due to high permittivity.



Gauss's law at target interface:

$$|E_{z\text{target}}| = \frac{|E_{z\text{plume}}|}{\epsilon_r} + \frac{\sigma}{\epsilon_0 \epsilon_r}$$



Electric field comparisons in dielectric BSO target

$V_P = +6$ kV, $t_f = 1.1$ μ s

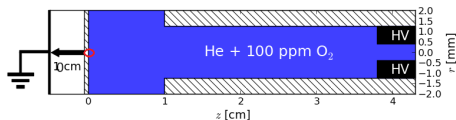
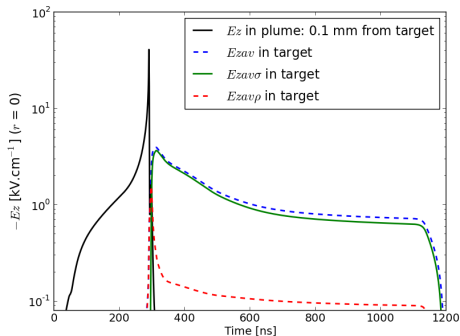
Why is the EF in the target lower than the EF in the plume?

Separation of the contributions to the EF calculation:

- EF inside the target with $\epsilon_r = 56$ due mostly ($\sim 90\%$) to high surface charge density (~ 30 nC \cdot cm $^{-2}$).
- Simulation results explain experimental measurements.

Gauss's law at target interface:

$$|E_{z\text{target}}| = \frac{|E_{z\text{plume}}|}{\epsilon_r} + \frac{\sigma}{\epsilon_0 \epsilon_r}$$



Review on physics of plasma jets and interaction with surfaces:

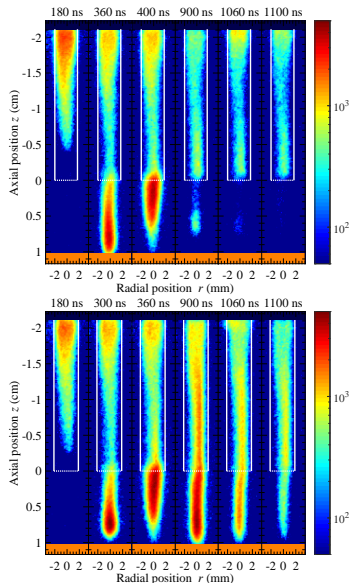
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Plasma jet interaction with metallic targets

Floating vs grounded target ($V_P = +5$ kV, $t_f = 1 \mu\text{s}$)

Imaging in jet in Eindhoven University of Technology, with rectangular pulse and different copper targets: **floating (top)** and **grounded (bottom)**.

- Return stroke with both targets.
- Luminous channel until end of pulse with grounded target but not floating.
- Additional emission near the source after end of pulse with floating target but not grounded.



P Viegas *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 095011

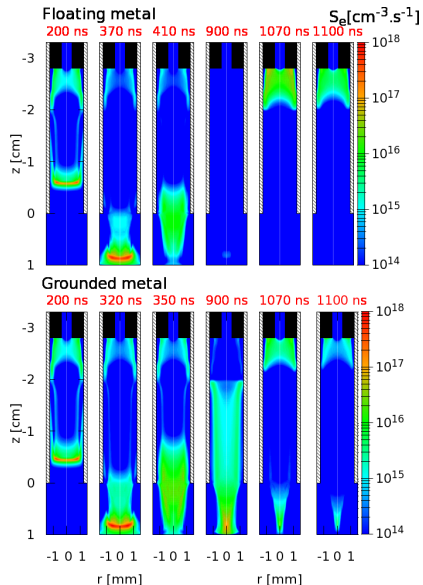
Plasma jet interaction with metallic targets

Floating vs grounded target ($V_P = +5$ kV, $t_f = 1 \mu\text{s}$)

Electron-impact ionization source term in simulations with different metallic targets: **floating and grounded**.

- Return stroke with both targets, as in experiments.
- EF in channel until end of pulse with grounded target but not floating.
- S_e near the source after end of pulse stronger with floating target than with grounded.

P Viegas *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 095011



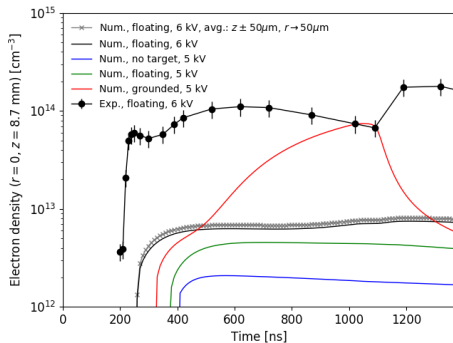
Electron density comparisons with metallic targets

Floating vs grounded target ($t_f = 1 \mu\text{s}$)

Electron density in plasma plume

measured through Thomson scattering with floating metallic target and simulated with different configurations (no target, grounded metal and floating metal).

- No agreement between simulations and experiments in n_e .
- Lack of knowledge on memory effects in model?
- Lack of knowledge on interaction between charges and metallic surfaces?
- Lack of benchmarking between experimental n_e diagnostics?



P Viegas et al., *Plasma Sources Sci. Technol.* 29 (2020) 095011

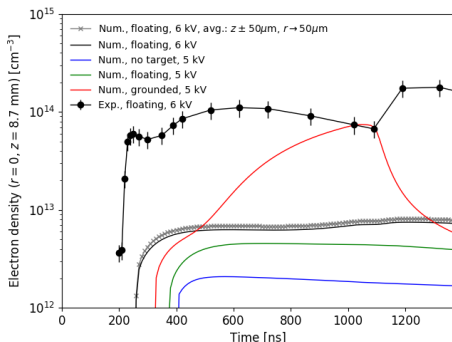
Electron density comparisons with metallic targets

Floating vs grounded target ($t_f = 1 \mu\text{s}$)

Electron density in plasma plume

measured through Thomson scattering with floating metallic target and simulated with different configurations (no target, grounded metal and floating metal).

- Higher n_e with return stroke than in free jet.
- Growth of n_e during the pulse with grounded target but not with floating.
Why?
- Growth of n_e after fall of pulse with floating target but not with grounded.
Why?



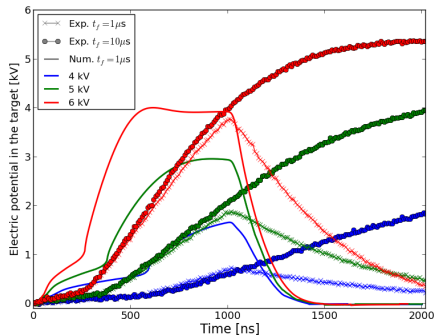
P Viegas et al., *Plasma Sources Sci. Technol.* 29 (2020) 095011

Plasma jet interaction with metallic targets

Charging of floating metallic target

Metallic target at floating potential charging when impacted by discharge and during pulse, and uncharging at the end of the pulse.

- No agreement between experiments and simulations.
- Target almost grounded when discharge arrives.
- Yet, target at HV at the end of the pulse.
- Explains similarities (return stroke) and differences (during and after pulse) in discharge dynamics with floating and grounded metallic targets.



P Viegas *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 095011

Insights from comparisons in jets with metallic targets

Summary

- A return stroke propagates **from near-grounded metallic targets to the powered electrode**.
- Ionization wave that partially neutralizes the net charge in the plasma channel, that propagates with reverse polarity with respect to the first ionization front.
- Floating target charges a few kV in μs timescale.
- Charge relaxation event after fall of pulse **between grounded source electrode and HV target**.
- Diffuse electrical redistribution that contributes to jet neutralization.

Characterization of plasma jets and interaction with targets

Conclusions

- Recent **quantitative, spatially and temporally-resolved comparisons of EF and n_e** between simulations and different experimental diagnostics in plasma jets.
- Reinforced confidence in key physical parameters of these plasmas, in particular the electric field - important for applications.
- Simulations used to **complement** experimental diagnostics. Provide detailed explanation of the dynamics of charges and EF in discharge propagation and plasma-target interactions.
- Comparisons show need for further understanding (disagreement in n_e and metallic target charging rates).

Review on physics of plasma jets and interaction with surfaces:

- Physical characterization of plasma jets
 - Numerical modelling
 - Diagnostics and experimental benchmarking
- Insights from comparisons between simulations and experiments
 - Jet interacting with dielectric target
 - Jet interacting with metallic targets
- Outlook on future of physics of plasma jets and interaction with surfaces

Outlook on future of physics of plasma jets and interaction with surfaces

- **Memory effects** between pulses are still an important unknown in physics of plasma jets (and other plasma sources).
 - Preionization, radicals, surface charges?
 - Dependence on conditions: polarity, target?
- Difficult to assess experimentally (low values) and numerically (long timescales).
- Simulation successfully describes surface charging memory effect by taking σ profile a few μs after the pulse from first simulation.
An (computationally) effective way to **obtain insight into memory effects**?
P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181
- Need new numerical and experimental methods?

Outlook on future of physics of plasma jets and interaction with surfaces

- Fluid simulations **describe complex (ns to ms) charging dynamics** of dielectric target, in agreement with experiments, by electric drift and simple boundary conditions (unit probability electron absorption and ion neutralization and s.e.e. with $\gamma_{se} = 0.1$).
Is it enough? Why disagreement charging metallic targets?
- Jets as repetitive stable transient discharges, ideal for surface studies.
- Need better description of sheath phenomena?
- Need microscopic description of **charge transfer between plasma and surface**?
- Increasing level of complexity of targets: rugosity, limited conductivity, liquid state.

Outlook on future of physics of plasma jets and interaction with surfaces

- Physical quantities (n_e) vary a lot with configurations: multiple diagnostics and comparisons with simulations in same system.
- **Quantitative comparisons** between measurements and simulations to increase confidence in critical parameters:
 - Sensitivity analysis.
 - Uncertainty quantification.
 - Verification and validation (V&V) methodologies.

Thank you for any feedback

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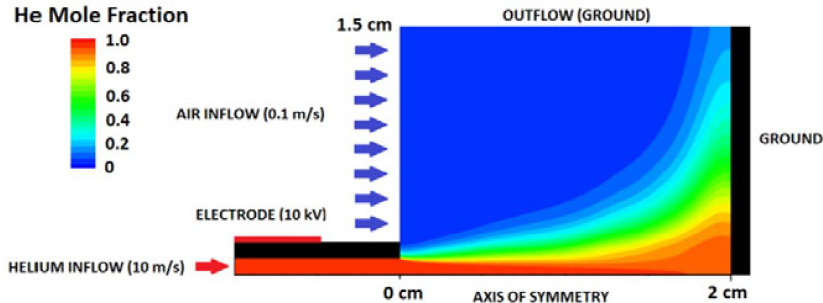
Acknowledgements:

HPC resources on cluster Hopper at École Polytechnique.

Project LM2018097 funded by Ministry of Education, Youth and Sports of the Czech Republic.

Project PTDC/FISPLA/1616/2021 (PARADiSE), funded by the portuguese FCT - Fundação para a Ciência e a Tecnologia.

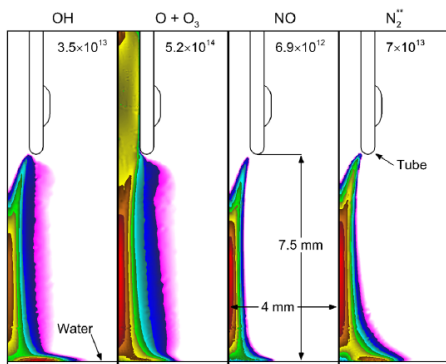
Plasma jets and interaction with targets



D Breden and L Raja, *Plasma Sources Sci. Technol.* 23 (2014) 065020

- Targets affect **gas mixing** in plasma plume between noble gas and air.
- Gas mixing affects distribution of densities and rate coefficients, with an effect on discharge path of propagation and interaction with targets.

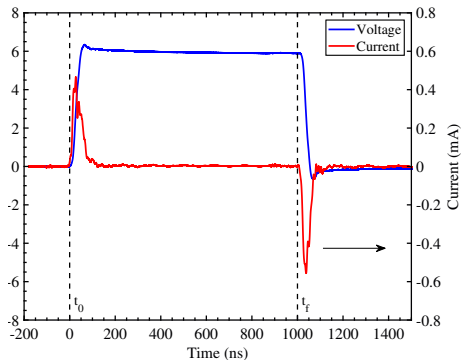
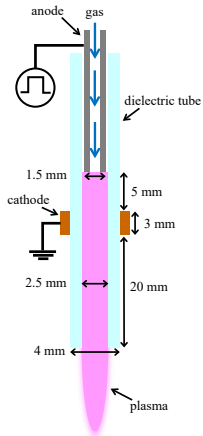
Plasma jets and interaction with targets



N Babaeva and M Kushner, *J. Phys. D: Appl. Phys.* 46 (2013) 025401

S Norberg *et al.*, *J. Phys. D: Appl. Phys.* 52 (2019) 015201

- Targets such as liquids and biological tissues induce complexity in chemistry in plasma plume.
- Models with hundreds of species and thousands of reactions.



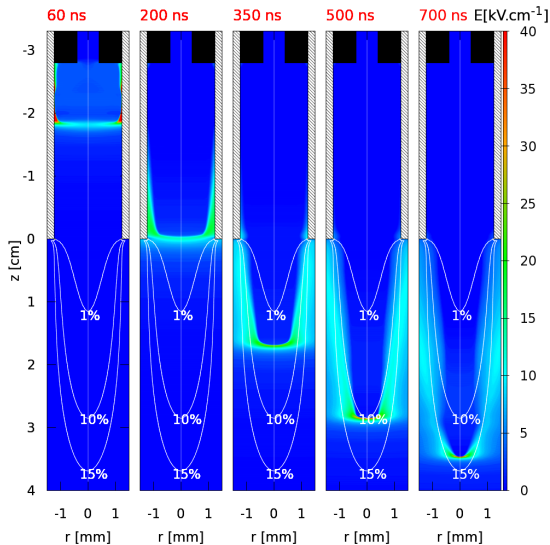
M Hofmans *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 034003

Discharge propagation in free jet

Discharge propagation in the tube and in the plume:

Simulated **spatial distribution of the magnitude of the EF at different instants** in agreement with experimentally obtained imaging:

- Peak EF in the front.
- Annular structure of propagation in the tube.
- Radial confinement in the plume.

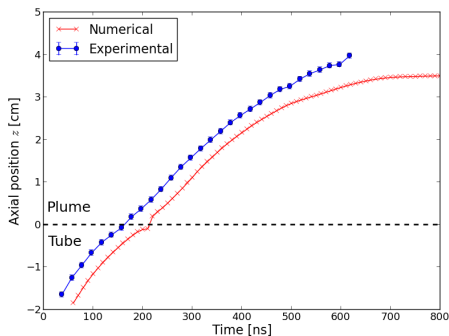


M Hofmans *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 034003

Discharge propagation in free jet

Position of the discharge front (maximum of EF or emission intensity) during axial propagation in the tube and in the plume.

- Same velocity in simulations and experiments. Of the order of $10 \text{ cm}\cdot\mu\text{s}^{-1}$ or $100 \text{ km}\cdot\text{s}^{-1}$.
- Different time of ignition. Uncertainty in memory effects at the source.
- Discharge stagnation during pulse.

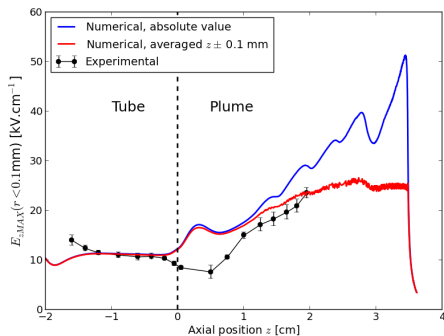


M Hofmans *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 034003

Electric field characterization in free jet

Peak EF during axial discharge propagation, simulated and measured through Stark-shift spectroscopy.

- High EF in front due to charge separation in volume.
- **Around $10 \text{ kV}\cdot\text{cm}^{-1}$ in tube and increasing in the plume, as in experiments.**
- Importance of averages taken in experiments.

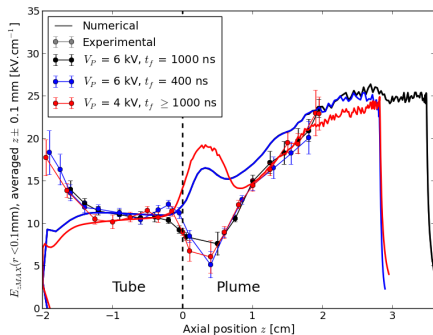
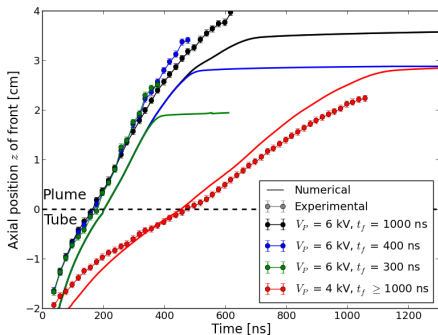


M Hofmans *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 034003

Discharge propagation in free jet

Changing magnitude and width of applied voltage:

- Larger discharge dimensions with higher magnitude.
- Faster propagation with higher magnitude.
- Earlier stagnation with shorter pulse.
- **Peak electric field independent.**



M Hofmans *et al.*, *Plasma Sources Sci. Technol.* 29 (2020) 034003

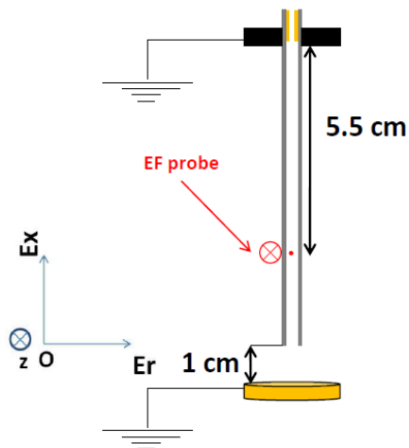
Plasma jets without target

Summary

- Qualitative agreement on discharge propagation and structure between simulations and imaging measurements.
- Excellent agreement on discharge propagation velocity in the tube and in the plume.
- High EF ($\sim 20 \text{ kV}\cdot\text{cm}^{-1}$) in the discharge front due to charge separation.
- Peak EF independent of magnitude of applied voltage.
- Importance of taking into account **spatial and temporal averages** when comparing simulation parameters with measurements.

Measurements of EF with grounded metallic target

T Darny et al., *Plasma Sources Sci. Technol.* 26 (2017) 045008



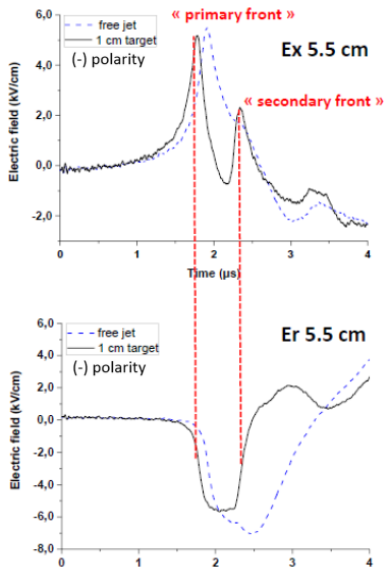
- Measurement of **time-resolved EF components** with an electro-optic probe at $r = 6$ mm:
Axial E_z and radial E_r .

Measurements of EF with grounded metallic target

T Darny *et al.*, *Plasma Sources Sci. Technol.* 26 (2017) 045008

With both polarities of applied voltage:

- Two peaks of E_z outside the tube.
- $|E_r|$ increases during the first peak and decreases during the second.



Plasma jet interaction with grounded metallic target

Discharge propagation and return stroke ($V_P = +6$ kV, different configuration)

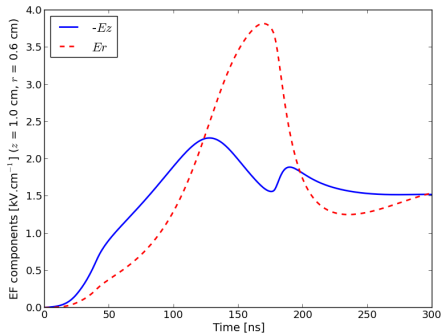
- Ring-shaped propagation inside the tube, with localized ionization front ($S_e \sim 10^{21} \text{ cm}^{-3} \cdot \text{s}^{-1}$). $S_e =$ electron-impact ionization source term.
- Impact on target at $t = 178$ ns. No spreading.
- Conductive plasma channel, with potential drop between electrodes.
- Diffuse propagation from target towards powered electrode.
- Return stroke as ionization wave ($S_e \sim 10^{19} - 10^{20} \text{ cm}^{-3} \cdot \text{s}^{-1}$).

Plasma jet interaction with grounded metallic target

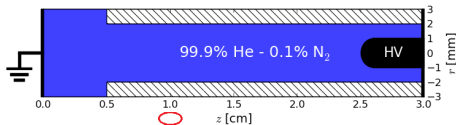
Return stroke ($V_P = +6$ kV), qualitative comparison

Temporal profiles of Electric Field components at $z = 1.0$ cm and $r = 0.6$ cm, outside the tube

- As in experiments, axial component E_z presents **two peaks**, before and after the front hits the target.
- As in experiments, radial component E_r **lowers after the** passage of the **return stroke**.

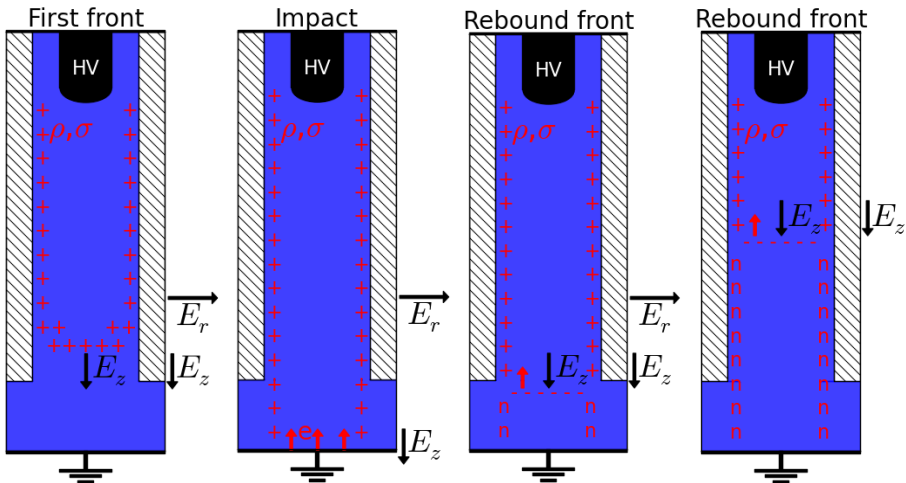


P Viegas et al. *Plasma Sources Sci. Technol.* 27 (2018) 025007



Plasma jet interaction with grounded metallic target

Return stroke as neutralization of the plasma channel



Plasma jet interaction with metallic targets

Summary

In agreement with experiments, with metallic targets:

- A return stroke propagates from the target to the powered electrode.
- Second peak of E_z and decrease of E_r outside the tube.

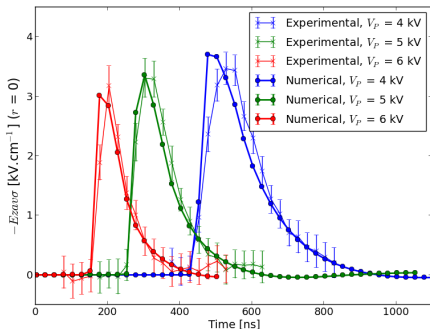
Simulation results complementary to experimental measurements:

- Cathode-emitted electrons contribute to sustaining return stroke.
- It propagates as an ionization wave that partially neutralizes the plasma channel, with reverse polarity with respect to the first ionization front.

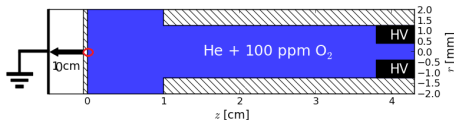
Electric field characterization in dielectric BSO target

Electric field inside the target.
Temporal profiles at $r = 0$, averaged through the target thickness.
Short pulses, with different magnitudes of applied voltage.

- Time of impact changes with magnitude of applied voltage.
- EF in target not dependent on magnitude of applied voltage, as in plume.



E Slikboer *et al.*, *Plasma Sources Sci. Technol.* 28 (2019) 095016

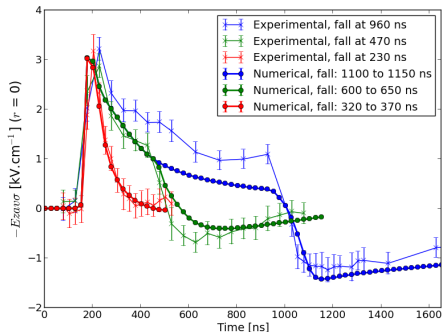


Electric field characterization in dielectric BSO target

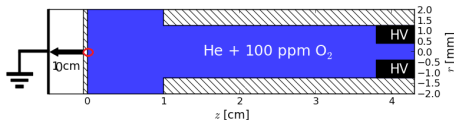
Electric field inside the target.
Temporal profiles at $r = 0$, averaged
through the target thickness.

$V_p = 6$ kV, different pulse lengths.

- During the pulse, as the discharge spreads radially, EF at $r = 0$ decreases.
- With long pulses and high charging, opposite polarity field after fall of pulse.



E Slikboer *et al.*, *Plasma Sources Sci. Technol.* 28 (2019) 095016



Electric field inside dielectric BSO target

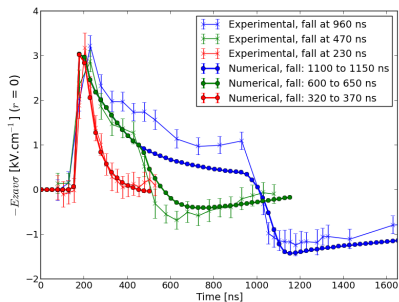
$$V_P = +6 \text{ kV}$$

Electric field inside the target, from **measurements and simulations**.

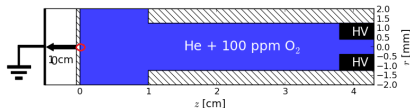
Temporal profiles at $r = 0$, averaged over the target thickness.

$V_P = +6 \text{ kV}$, different pulse widths.

- During the pulse, positive charging.
- As the discharge spreads radially, E_z at $r = 0$ decreases.
- With long pulses and high charging, opposite field after fall of pulse.
- **Charging time** determines interaction: control by distance, applied voltage and pulse width.



E Slikboer *et al.*, *Plasma Sources Sci. Technol.* 28 (2019) 095016



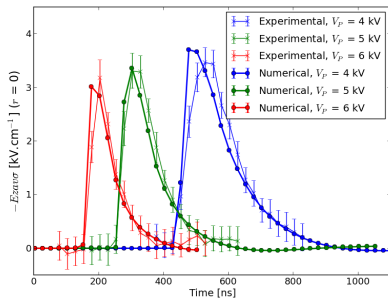
Electric field inside dielectric BSO target

Short pulses

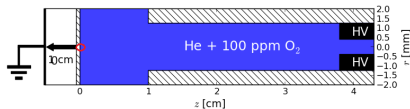
Electric field inside the target.
Temporal profiles at $r = 0$, averaged
through the target thickness.

Short pulses, with different magnitudes of
applied voltage.

- Time of impact changes with
magnitude of applied voltage.
- EF in target not dependent on
magnitude of applied voltage, as in
plume.



E Slikboer *et al.*, *Plasma Sources Sci. Technol.* 28 (2019) 095016



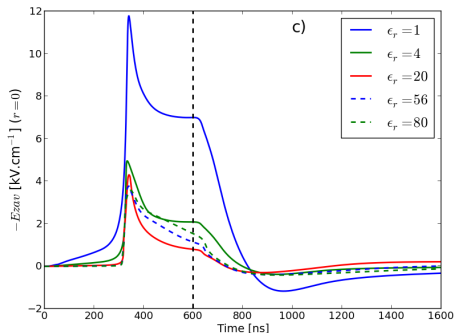
Electric field comparisons in dielectric BSO target

$$V_P = +6 \text{ kV}, t_f = 0.6 \mu\text{s}$$

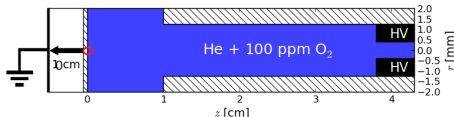
Is the EF measured inside BSO targets representative of the EF inside any dielectric target with impinging jet?

Simulations of jets interacting with dielectric targets of different permittivities:

- For floating targets with $\epsilon_r \geq 4$, similar simulated $E_z(t)$ at $r = 0$.
- Simulation results complement experimental measurements that cannot use every dielectric material.



P Viegas and A Bourdon, *Plasma Chem. Plasma Process.* 40 (2020) 661-683



Insights from comparisons in jets with dielectric targets

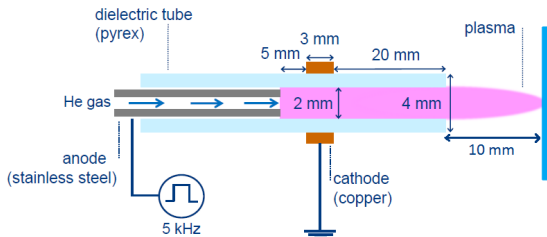
Summary

- Discharge spreading and EF penetration within dielectric targets.
- **Quantitative comparison of EF inside BSO target** between simulations and experiments with excellent agreement.
- The **EF** inside the target **has a relatively low magnitude ($\sim 3 \text{ kV}\cdot\text{cm}^{-1}$)**, in both **simulation and measurements** through Mueller polarimetry.
EF in target lower than in plume.
- Dielectric targets of high ϵ_r shield the EF from the plasma and are exposed to **EF** generated **mostly by σ** deposited on the surface.
- EF in BSO targets ($\epsilon_r = 56$) is representative of floating dielectric targets with $\epsilon_r > 4$.

Jet experiments with dielectric BSO target

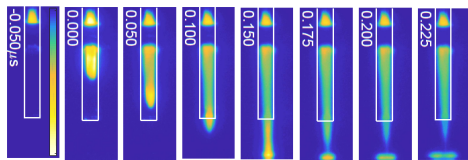
E Slikboer, *PhD thesis* (2018)

- Jet generated by rectangular $V_P = +6/-6$ kV pulse (e.g. $t_f = 1 \mu\text{s}$ width) applied to electrode inside the tube with $f = 5$ kHz.
- Flow of 1.0 L/min of Helium through the pyrex tube into air atmosphere.
- Interaction between plasma plume and floating dielectric BSO (electro-optic) crystal ($\epsilon_r = 56$ and 0.5 mm thickness) at 1 cm from the end of the tube.



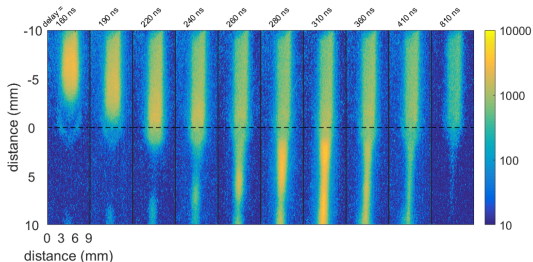
Jet experiments with dielectric BSO target

+6 kV



E Slikboer, *PhD thesis* (2018)

-6 kV



P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181

Imaging results:

- Discharge impact and spreading on the target.
- Slower propagation for **negative polarity**.
- Formation of **charged cloud on target before impact**: potential memory effect?
- Discharge reconnection rather than impact.

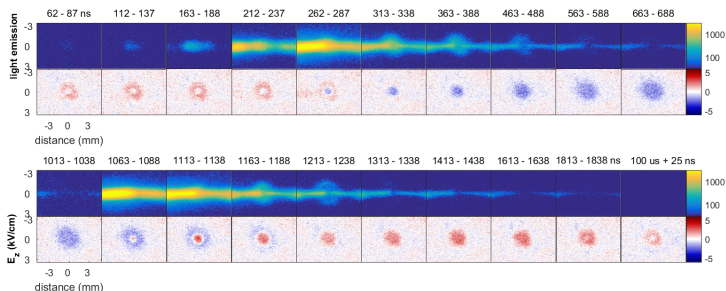
Simulation of plasma jet with dielectric target

Electron-impact
ionization source term S_e
during the pulse,
from $t = 0$ to $t = 800$ ns

- Discharge propagation, impact and spreading on the target.
- Slower propagation for negative polarity.
- As in experiments, **charged cloud on target before impact, due to initial surface charge.**
- Discharge reconnection rather than impact.
- Why this memory effect for negative and not positive polarity?

Electric field inside dielectric BSO target

$V_P = -6$ kV and $t_f = 1010$ ns



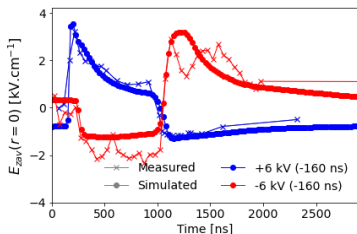
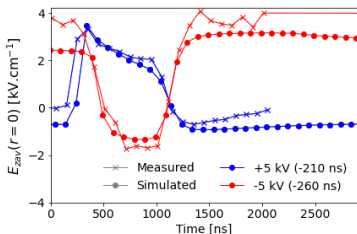
P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181

Imaging and Mueller polarimetry measurements of E_z inside BSO target.
Spatial profiles of E_z at different times for $V_P = -6$ kV.

- As expected, in the opposite sense of the +6 kV case, negative charging during pulse and positive charging after the fall of the pulse.
- Unlike positive case, **significant surface charging remaining between pulses**, between $t = 2 \mu$ s and $t = 200 \mu$ s (next pulse)

Electric field inside dielectric BSO target

$t_f = 1010$ ns



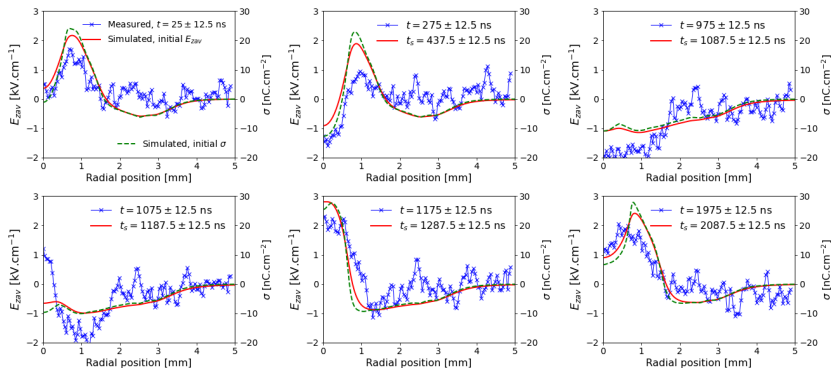
P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181

Measured and simulated $E_z(r=0)(t)$ inside BSO target for $V_p = +5/-5$ kV and $+6/-6$ kV, averaged axially and temporally.

- Agreement between simulations and experiments.
- Positive E_z and charge remaining on the target surface in negative polarity cases.

Electric field inside dielectric BSO target

$V_P = -6$ kV and $t_f = 1010$ ns



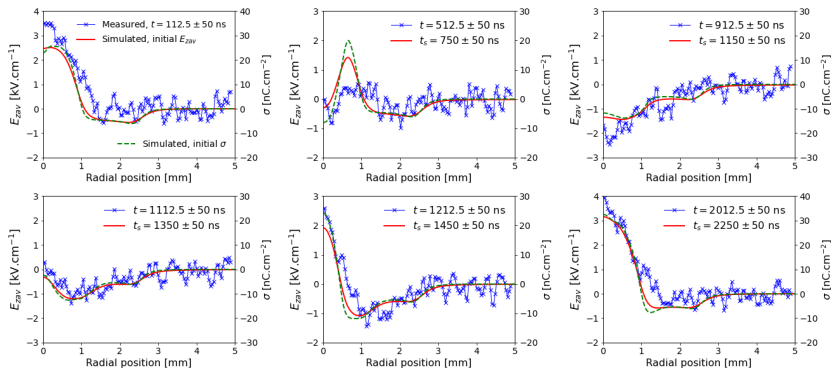
P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181

Measured and simulated $E_z(r)$ and σ at different times inside BSO target for $V_P = -6$ kV, averaged axially and temporally.

- Agreement between simulations and experiments.
- Positive E_z and σ remaining on the target with maximum around $r = 1$ mm. Why?

Electric field inside dielectric BSO target

$V_P = -5$ kV and $t_f = 1010$ ns



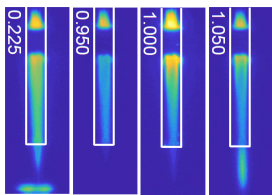
P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181

Measured and simulated $E_z(r)$ and σ at different times inside BSO target for $V_P = -5$ kV.

- Agreement between simulations and experiments.
- Positive E_z and σ remaining on the target, even at $r = 0$.

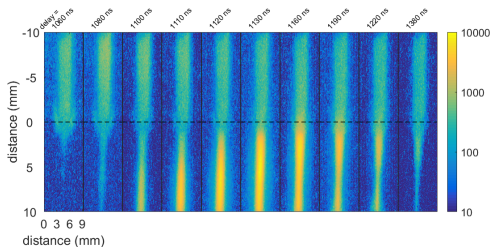
Dynamics and charging after the pulse

+6 kV



E Slikboer, *PhD thesis* (2018)

-6 kV



P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181

Experimental imaging results:

- For both polarities, luminous charge relaxation event between newly-grounded inner electrode and highly charged target.
- Leads to opposite-polarity charging of the target.

Dynamics and charging after the pulse

Surface charging memory effect

Simulated electric field magnitude E_t
and surface charge density σ
from $t = 1 \mu\text{s}$ to $t = 2.5 \mu\text{s}$

- Charge relaxation event between newly-grounded inner electrode and highly charged target leads to opposite-polarity charging.
- Afterwards, electric field remains between charged target and neutral post-discharge and target partially neutralizes.
- Why is there no charge drift and **no full neutralization of the target?**

Dynamics, charging and surface memory effects after pulse

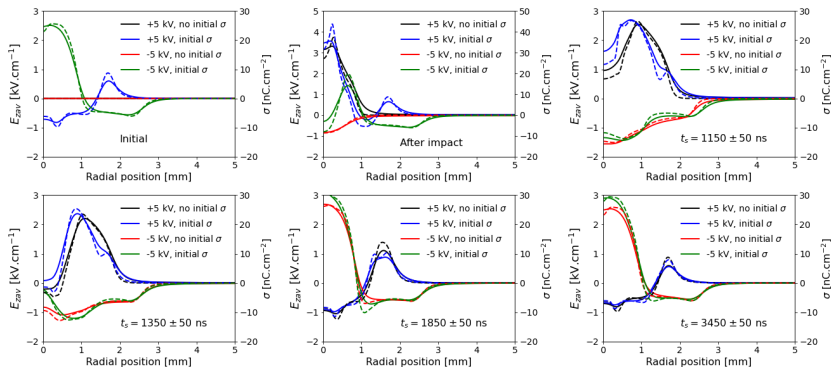
Surface charging memory effect

Simulated electron density n_e
and surface charge density σ
from $t = 1 \mu\text{s}$ to $t = 3.5 \mu\text{s}$

- Some time after the pulse, **free charges lack in the plasma** close to the target.
- Neutralization process stops.
- More pronounced for negative polarity.
- Hypothesis from simulation results may explain **surface charging memory effect for negative polarity**.

Electric field inside dielectric BSO target

$V_P = +5/-5$ kV and $t_f = 1.2$ μ s



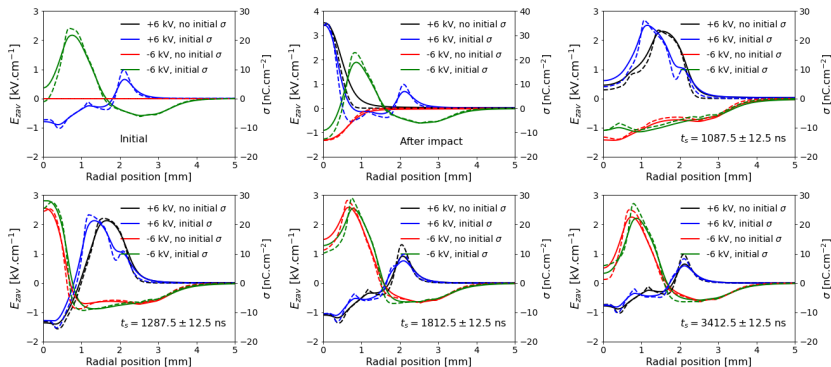
P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181

Simulated $E_z(r)$ and σ at different times inside BSO target for $V_P = +5/-5$ kV.

- E_z and σ remaining on the target for negative polarity more than for positive polarity.
- Effect of initial σ on impact. Convergence after the pulse.

Electric field inside dielectric BSO target

$V_P = +6/-6$ kV and $t_f = 1.1$ μ s



P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181

Simulated $E_z(r)$ and σ at different times inside BSO target for $V_P = +6/-6$ kV.

- E_z and σ remaining on the target for negative polarity more than for positive polarity.
- Effect of initial σ on impact. Convergence after the pulse.

Effect of jet polarity on charging of a dielectric target

Summary

- Charging of dielectric BSO target for both polarities (during the pulse and after the pulse).
- **Quantitative comparison of EF inside BSO target** between simulations and experiments with excellent agreement: validation and confidence.
- **EF and surface charge remain on target** in between pulses (200 μ s) for negative polarity only: important for applications.
- **Surface charging memory effect** for negative polarity demonstrated to affect discharge dynamics.
- Hypothesis from simulation results: neutralization of target after pulse dependent on availability of free charges in the plasma.

P Viegas *et al.*, *Sci. Rep.* 12 (2021) 1181