INTERACTIONS BETWEEN ATMOSPHERIC PRESSURE PLASMAS AND COMPLEX SURFACES*

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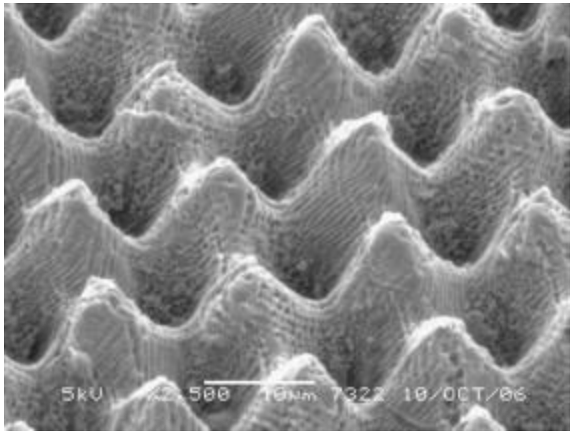
AGENDA

- Synergistic interactions with complex surfaces
- Primer on Surface Ionization Waves
- Description of model
- Surface discharges across wavy and aerosol hosting surfaces
- Surface discharges across porous materials.
- Plasma jets incident onto shallow, wide and deep channels.
- Concluding Remarks

PLASMA SYNERGIES WITH COMPLEX SURFACES



- **Atmospheric pressure plasmas (APPs) synergistically interact with complex surfaces.**
- **In this talk, complex means non-planar, non-uniform, porous dielectrics (wet and dry).**
- **Just as with metals, non-planar dielectrics having large ε_r produce electric field enhancement that intensify the plasma.**
- **Unlike metals, dielectric have memory through charging of their surfaces.**
- **Charging (and secondary electron emission) produce synergistic effects.**
- **As in low pressure plasmas, aspect ratio of features determine plasma penetration and properties.**

https://fineartamerica.com/featured/1-water-drops-on-a-rubber-surface-daniel-sambrausscience-photo-library.html

www.utwente.nl/en/et/ms3/research-chairs/WAoud_niets_uit_wissen_aub/research/laser/probic/#resultsjects_archive/hydropho

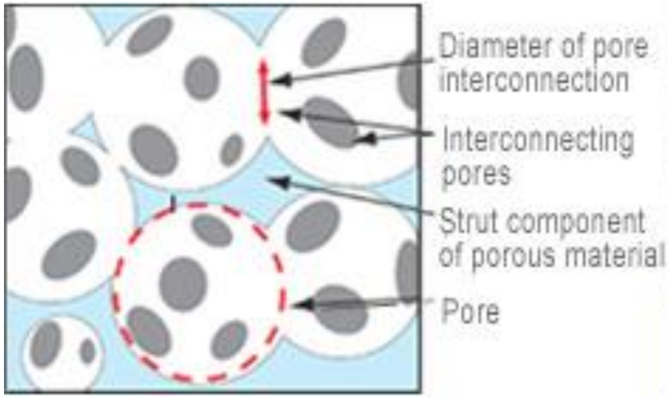
PLASMA TREATMENT OF BONE SCAFFOLDING

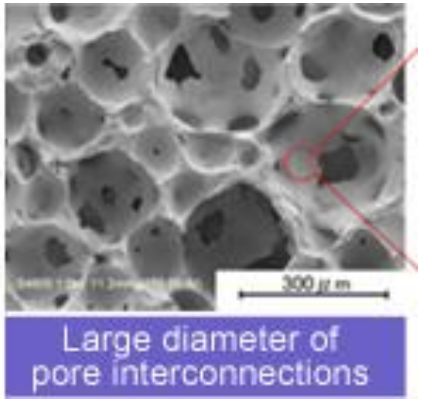
- **Bone scaffolding – calcium dominated, porous dielectric used as bone replacement. Bone cells grow into pores, eventually replacing scaffold.**
- **Scaffolding often needs treatment to improve biocompatibility.**

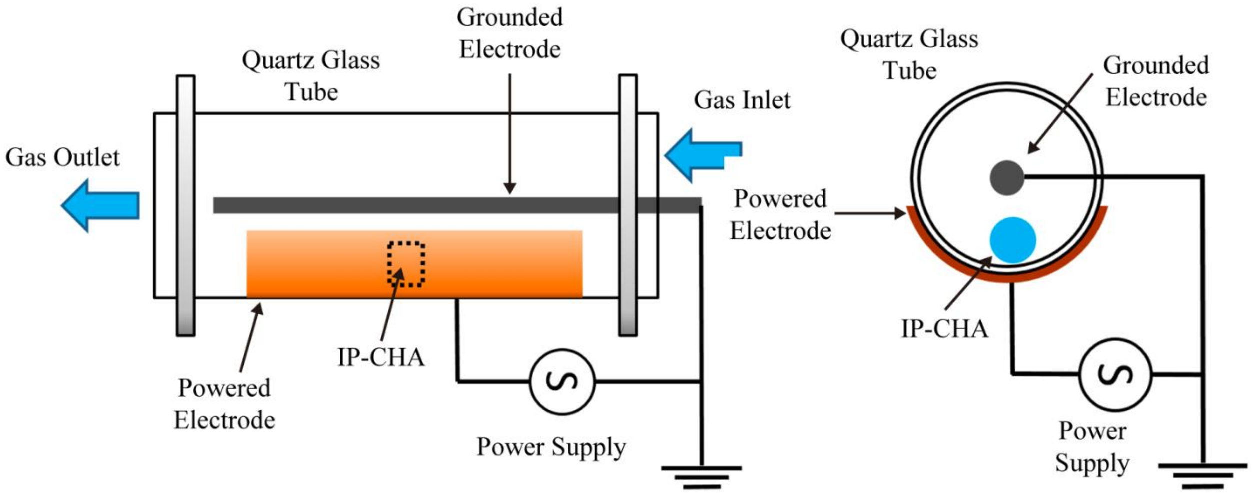
Impact of non-thermal plasma surface modification on porous calcium hydroxyapatite ceramics for bone regeneration

Yu Moriguchi , Dae-Sung Lee , Ryota Chijimatsu , Khair Thamina , Kazuto Masuda , Dai Itsuki , Hideki Yoshikawa , Satoshi Hamaguchi , Akira Myoui

PLOS ONE | https://doi.org/10.1371/journal.pone.0194303 March 14, 2018







www.coorstek.co.jp/eng/products/bio/neobone.html

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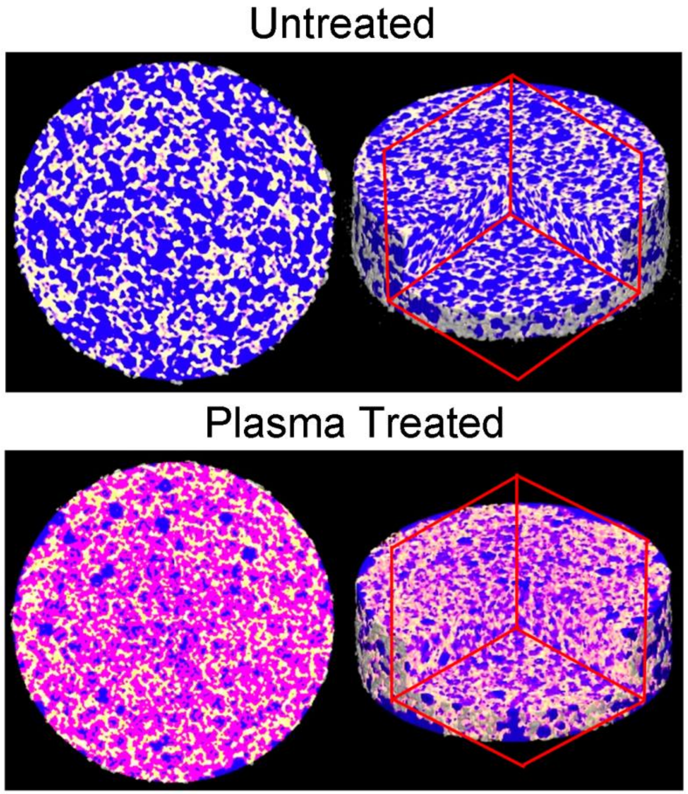
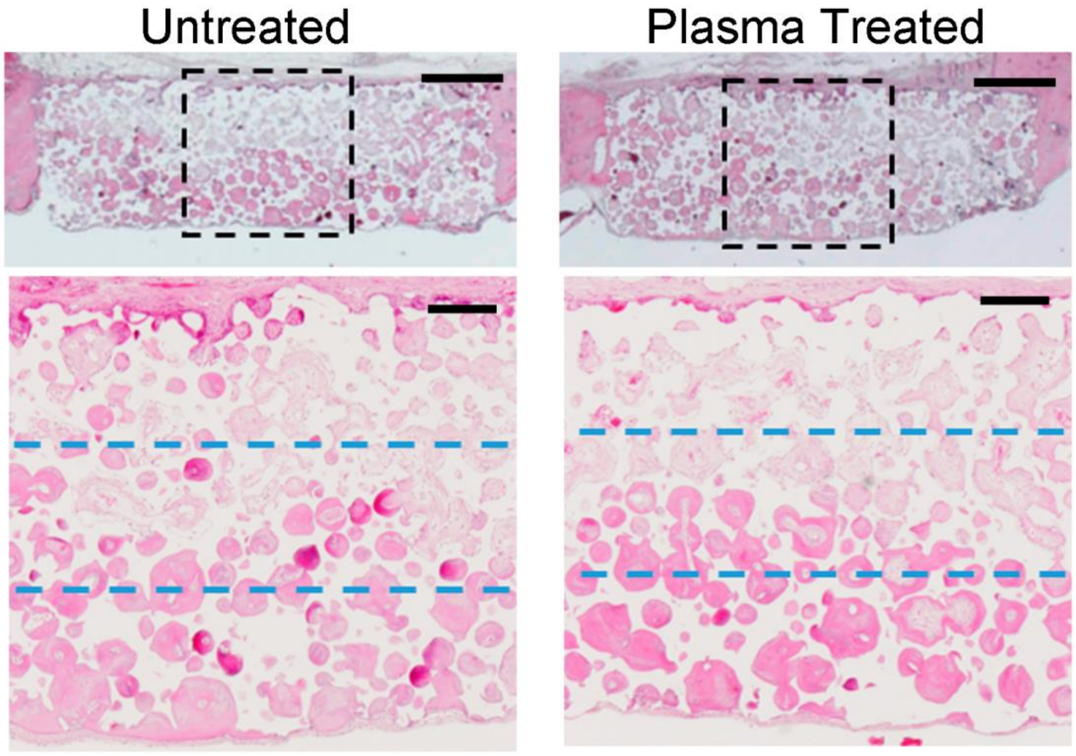
PLASMA TREATMENT OF BONE SCAFFOLDING

- **Low pressure oxygen plasma treatment of NeoBone (150 μm pores, 40 μm opengings) increases contract angle, wettability and cell proliferation into scaffolding.**
- **Can such processes be performed faster and cheaper using APPs?**





- **Wettability**
- **Cell proliferation**

- **Y. Moriguchi, et al. PLOS One, 2018**

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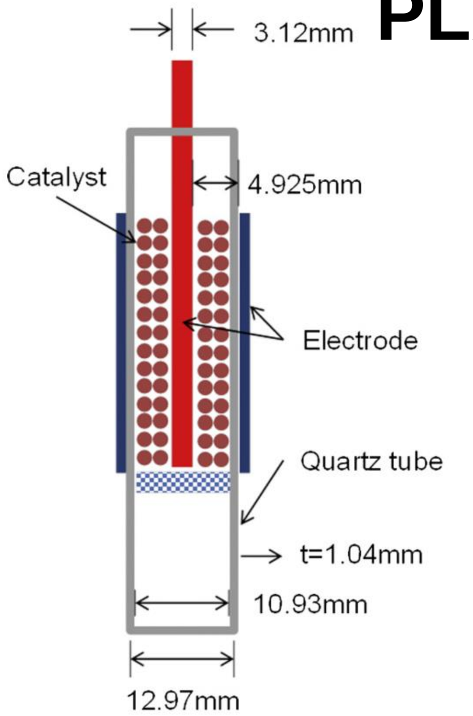
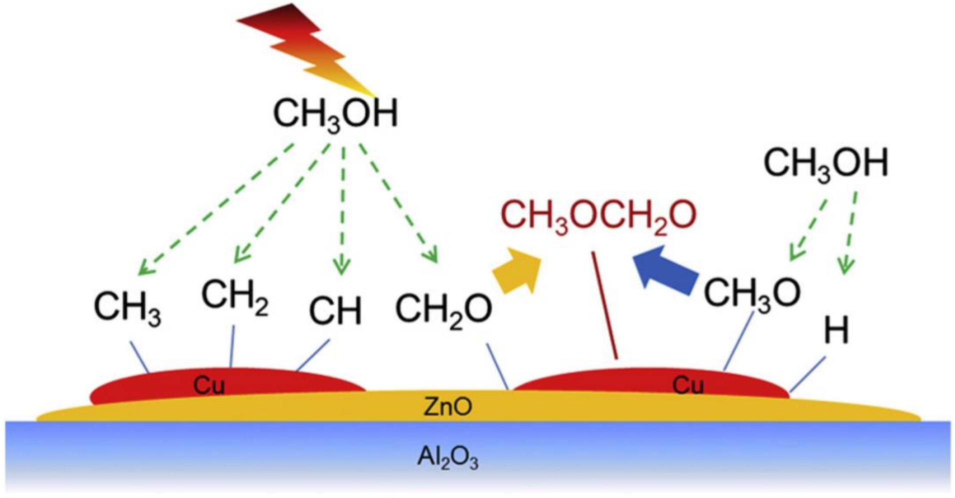
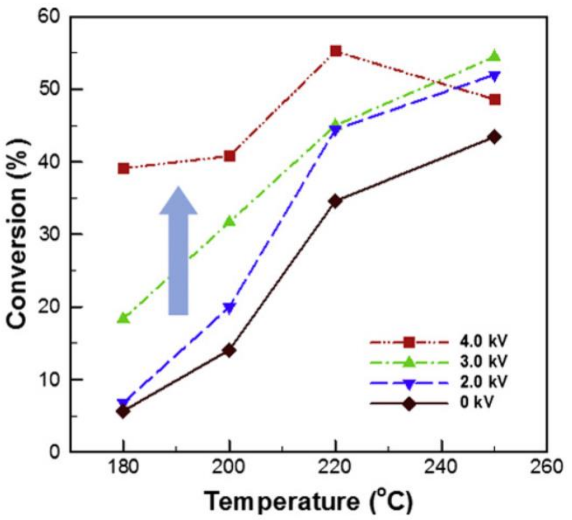
PLASMA CATALYSIS: CH_3OH REFORMING



- Packed bed plasma reactor with Cu/ZnO on Al_2O_3 catalyst improves the efficiency of methanol reforming.
- Higher conversion at lower temperature enabled by plasma formed radicals.
- Poorly understood interactions with porosity of dielectrics.





Synergetic mechanism of methanol–steam reforming reaction in a catalytic reactor with electric discharges
Taegyu Kim , Sungkwon Jo , Young-Hoon Song , Dae Hoon Lee
Applied Energy 113 (2014) 1692–1699

University of Michigan
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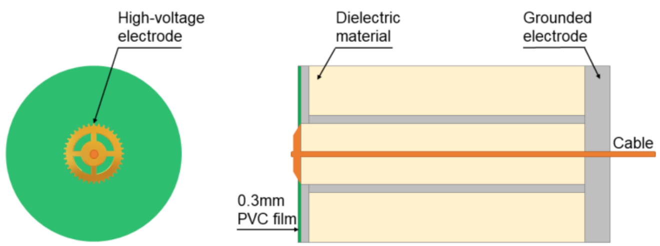
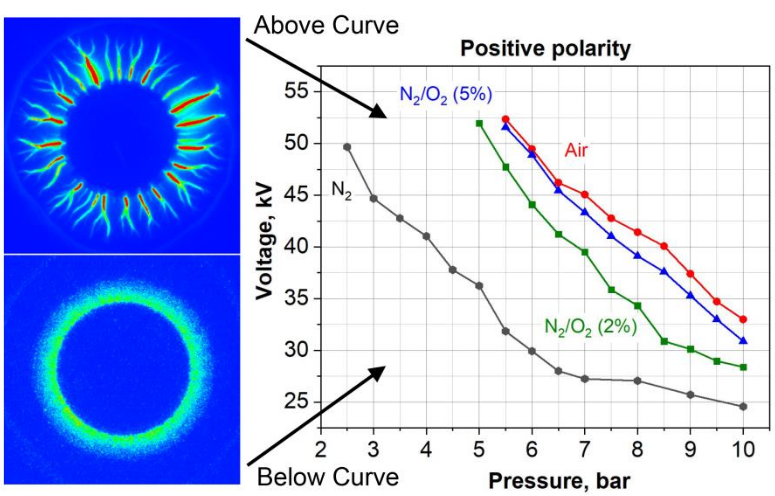
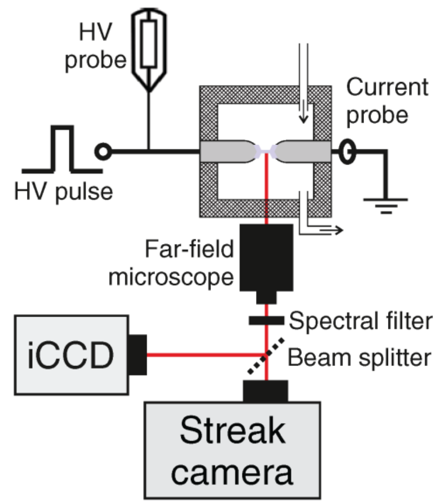
APPs IN CONTACT WITH SURFACES

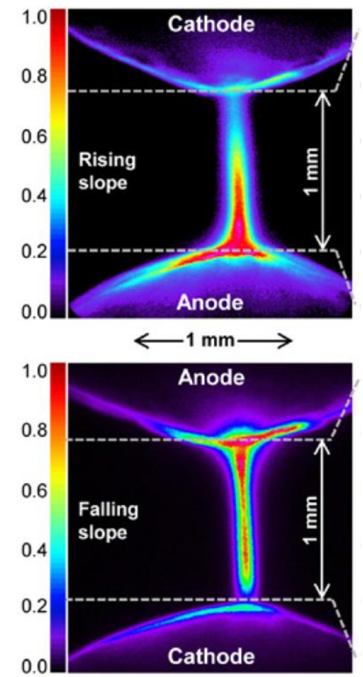
- **The majority of these applications involve direct contact of APPs with surfaces (solid, soft, liquid).**
- **One mode of such interactions involve “surface hugging” plasmas or “surface ionization waves” (SIWs) – either deliberate or natural outcome.**





- **Modes of Deliberate SIW (S. Starikovskaia, AIAA Scitech 2019 Forum)**





- **Natural transition to SIW (Hoft and Brandenburg, JPD 47, 465206 (2014).**

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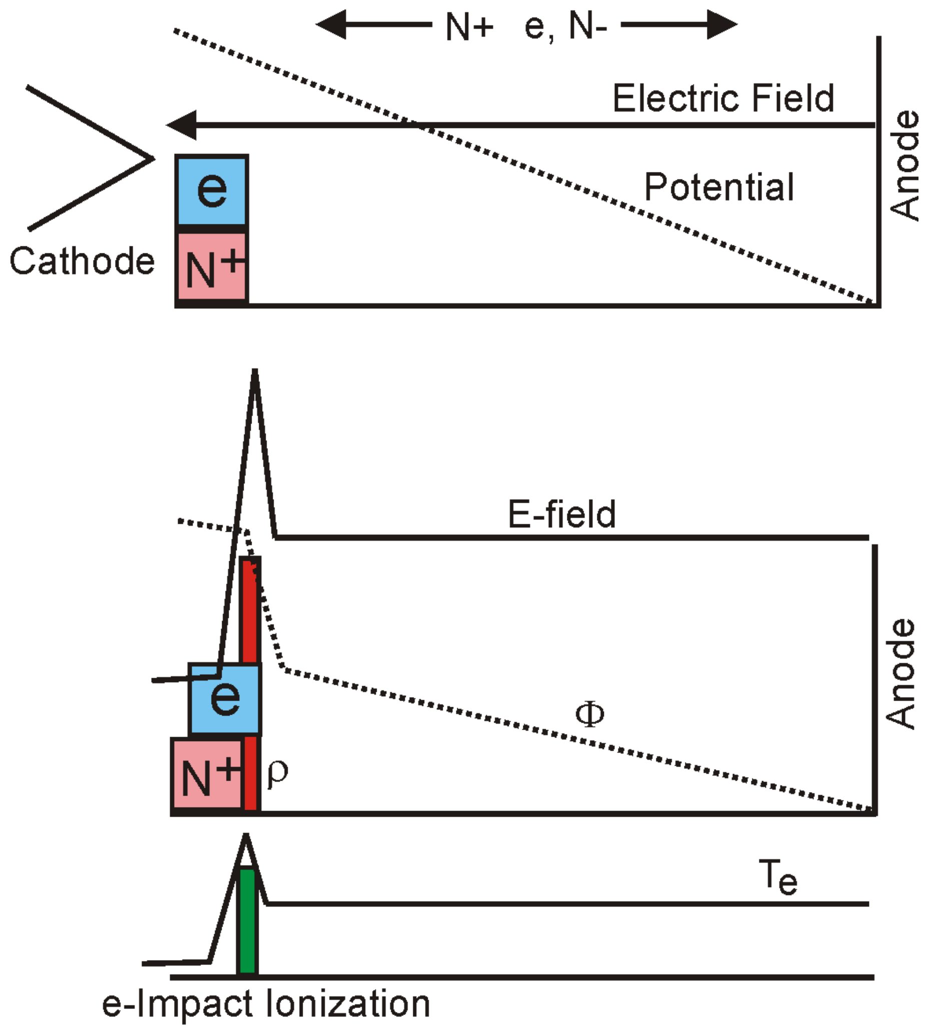
PRIMER ON SURFACE IONIZATION WAVES

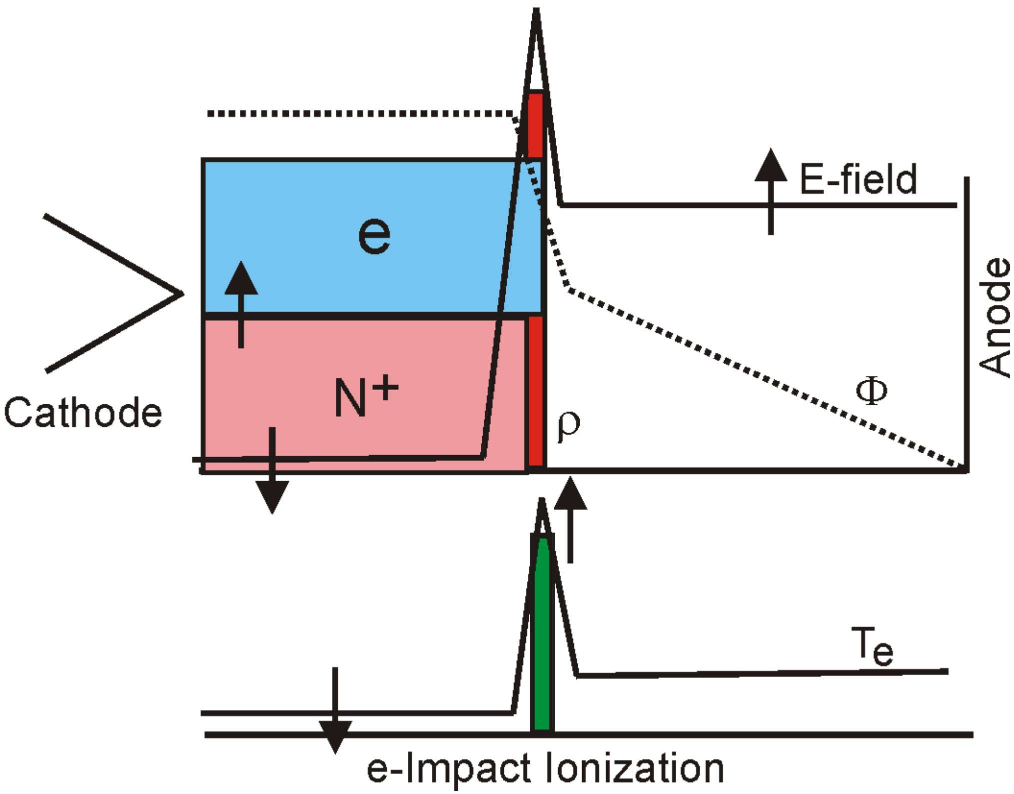
FUNDAMENTALS OF NEGATIVE "BULK" IONIZATION WAVES

- **Large negative voltage on cathode – grounded anode.**
- **Small spot of plasma at tip of cathode (field emission?).**
- **Electrons drift towards anode – ions towards cathode.**
- **Negative space charge creates spike in E-field.**
- **E-field heats electrons producing spike in T_e and ionization.**
- **Avalanche front develops, increasing plasma density.**



FUNDAMENTALS OF NEGATIVE STREAMERS

- **Avalanche front produces a column of high plasma density.**
- **High conductivity in column reduces E-field, voltage drop and T_e.**
- **Voltage compressed ahead of streamer, increasing E.**
- **Electrons drift towards anode – ions towards cathode.**
- **Negative space charge creates spike in E-field.**
- **E-field produces spike in T_e and ionization – avalanche front extends plasma column.**



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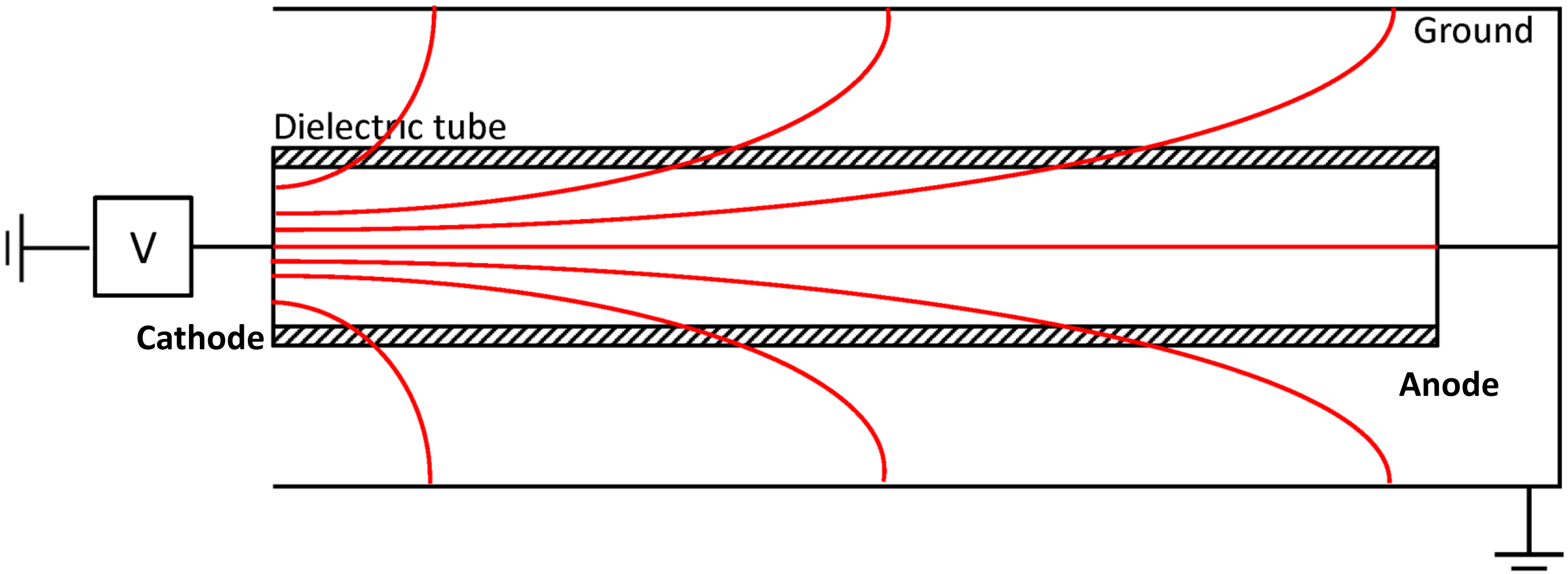
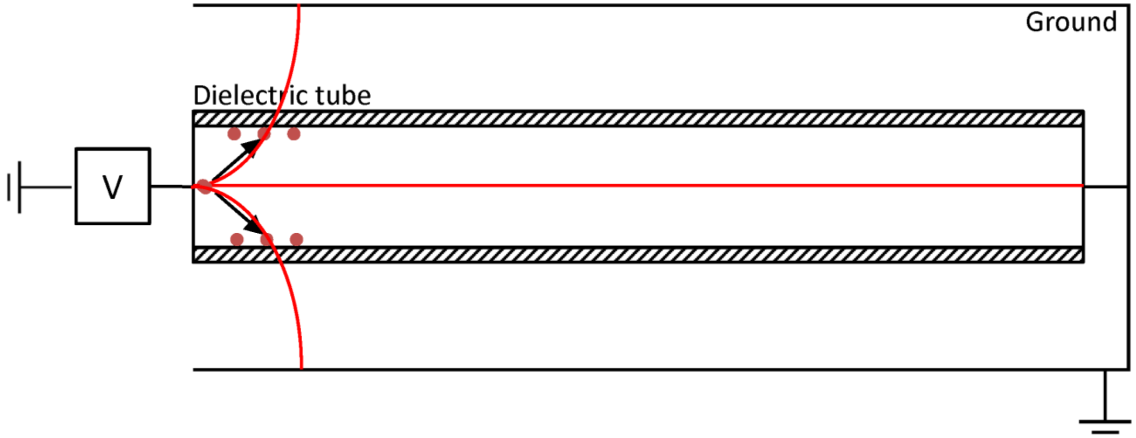
BREAKDOWN PROCESS – INITIAL E-FIELD

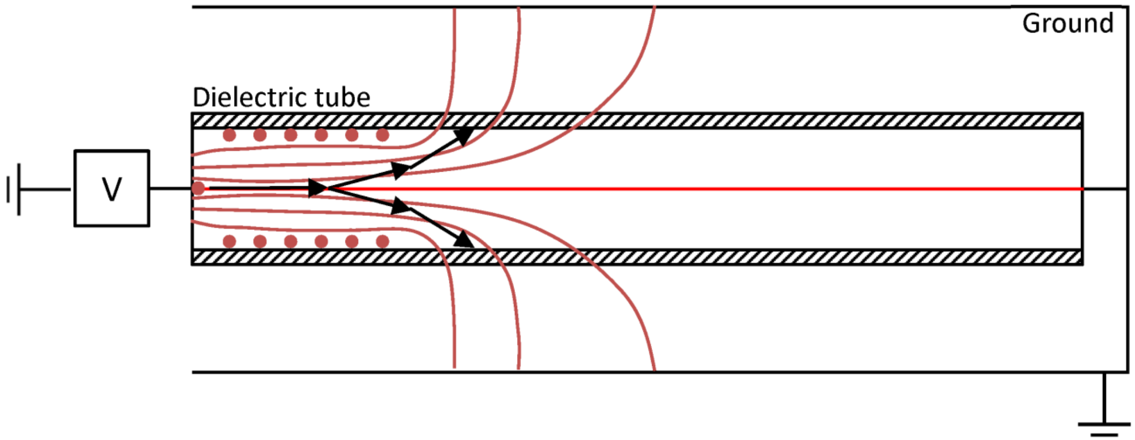
- Consider electrical breakdown in a fluorescent lamp-type discharge sustained in a dielectric tube.
- Initially, electric field lines are directed from cathode to *where-ever ground can be found*, here to the coaxial ground.
- The closest ground plane is rarely the anode – only electric field lines near the axis terminate on the anode



BREAKDOWN PROCESS – REDIRECTING E-FIELD

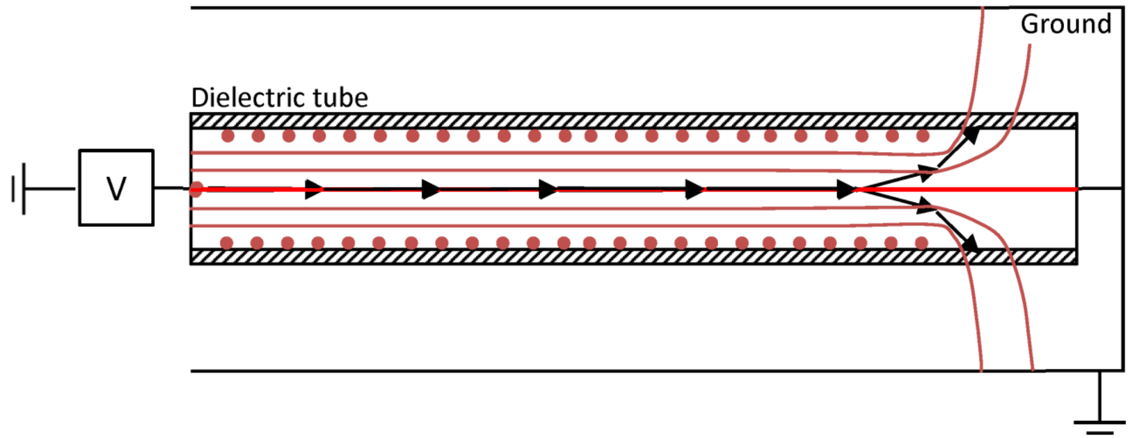
- Initial electrons from cathode follow vacuum electric field lines which intersect walls.
- Electrons charge wall, which shields potential and redirects E-field parallel to wall.
- Electrons drift axially to end of charged region, where they follow E-field to further change wall.

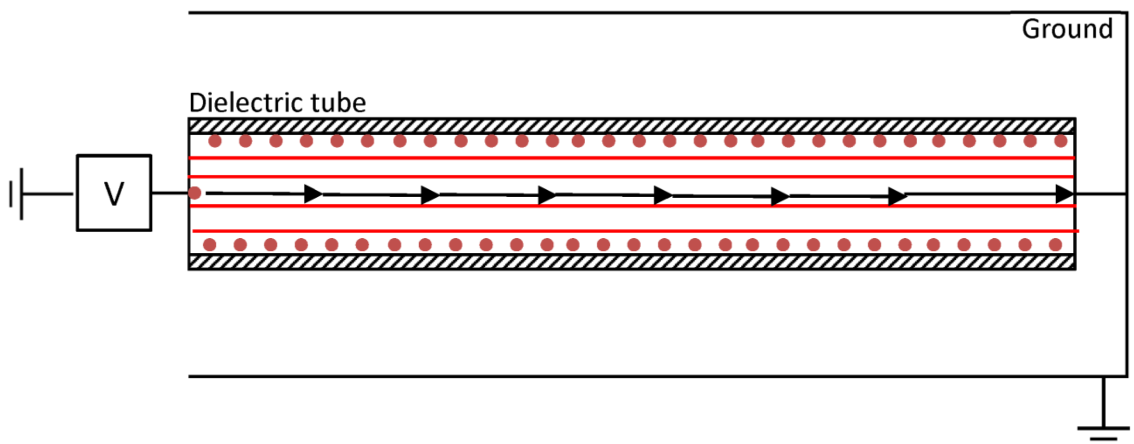




BREAKDOWN PROCESS – CONNECTING TO ANODE

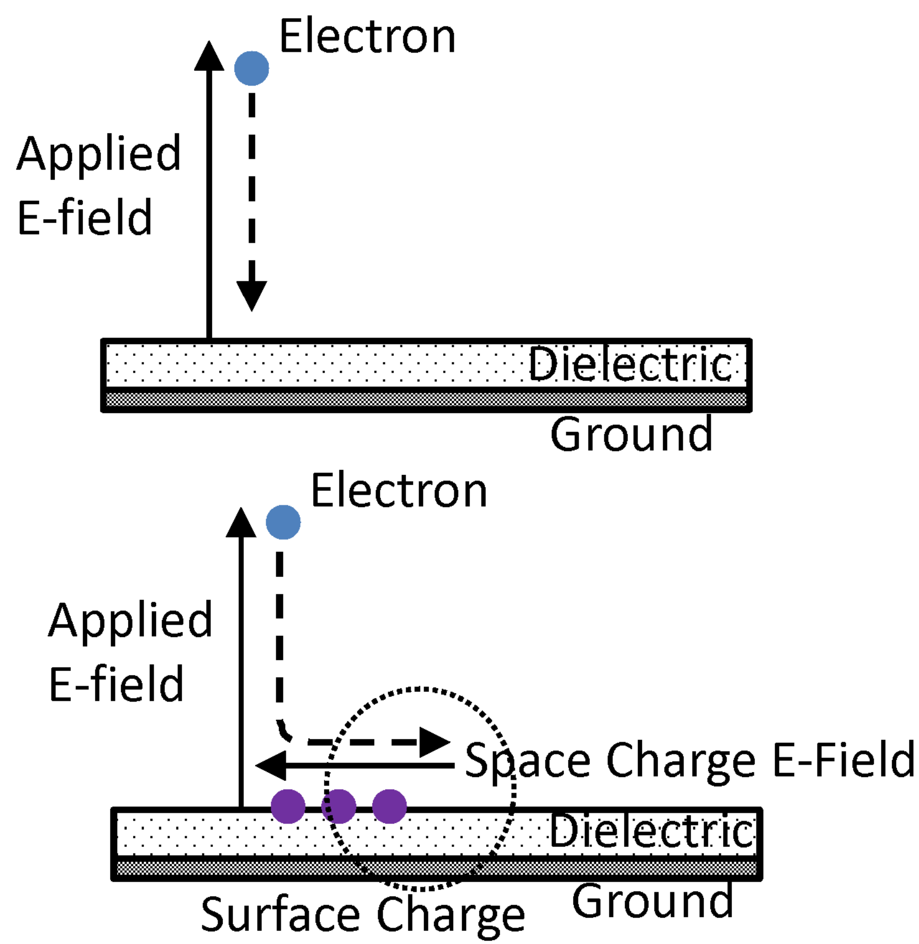
- Process of wall charging, turning E-field parallel to wall and electrons further charging wall continues along length of tube.
- Eventually, E-field lines seek out the cathode is closest ground plane.
- With full wall charging, coaxial ground is shielded from plasma.





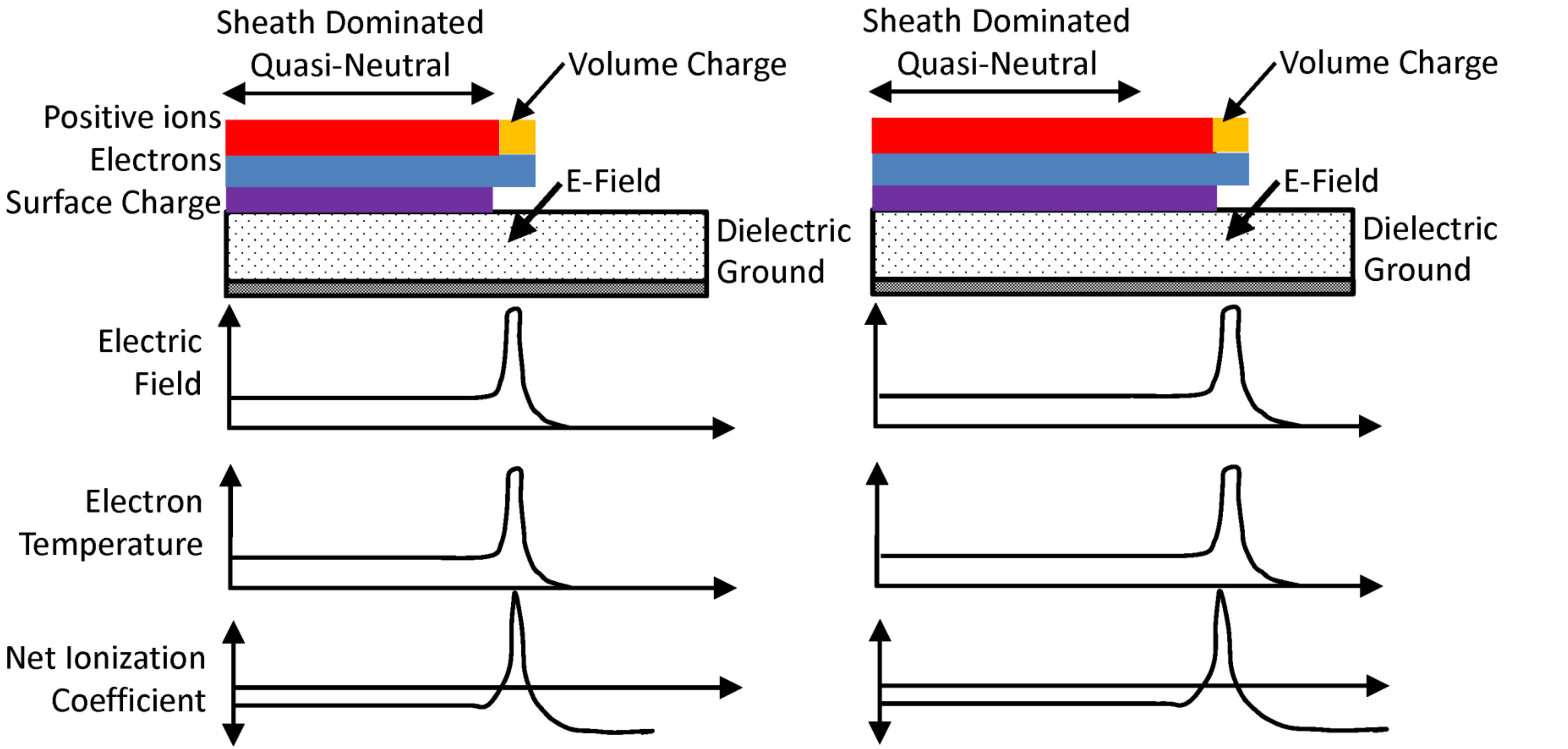
SURFACE IONIZATIONS WAVES

- **Surface Ionization Waves (SIWs) are streamer-like extensions of the plasma column that *hug* the dielectric.**
- **Space charge enhancement at the front of the SIW produces electric fields that avalanches the gas.**
- **Surface charging produces a sheath that shields underlying ground.**
- **Volume charge in head of stream produces peak in T_e.**
- **For negative SIW, process starts with external electric field driving electrons into surface,**



SURFACE IONIZATIONS WAVES

- **Electric field in surface-hugging plasma consumes voltage.**
- **SIWs will continue to propagate as long as there is sufficient voltage remaining.**



E-FIELD AND POTENTIAL: DIELECTRICS

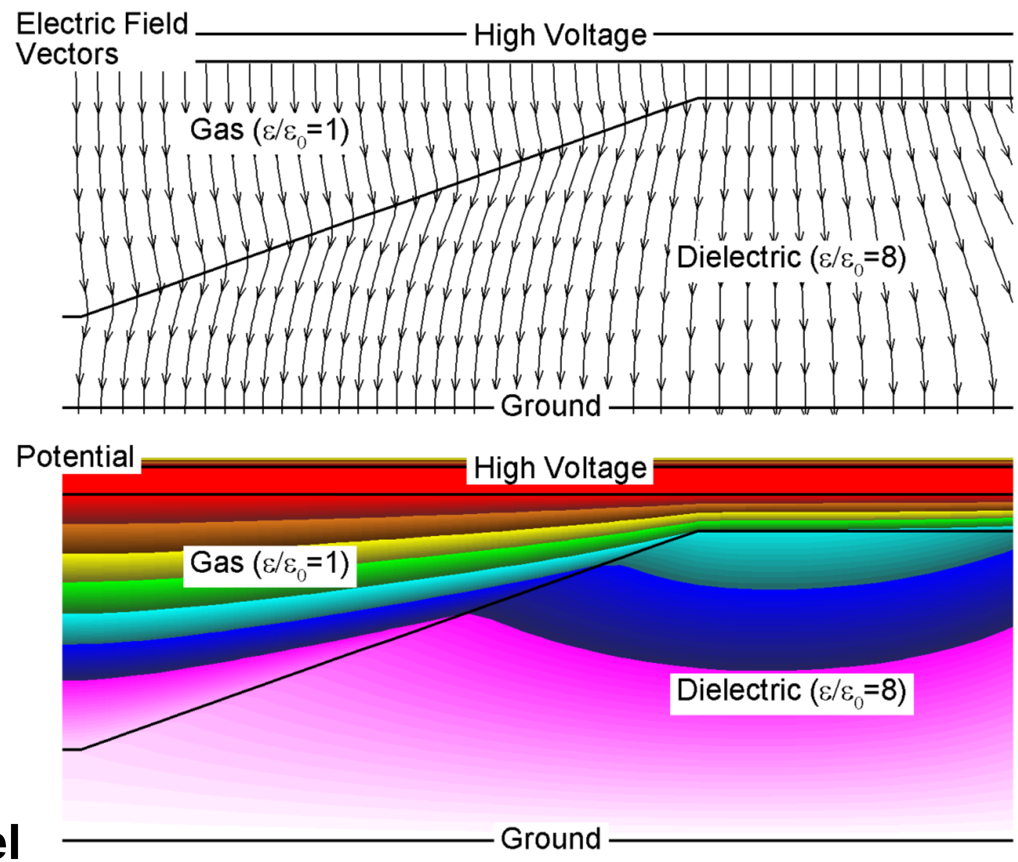
- **At surface of a dielectric and a gas:**

$$D_{\perp gas} - D_{\perp dielectric} = \rho_{surf}$$

$$\varepsilon_g E_{\perp gas} = \varepsilon_d E_{\perp dielectric} \ (\rho_{surf} = 0)$$

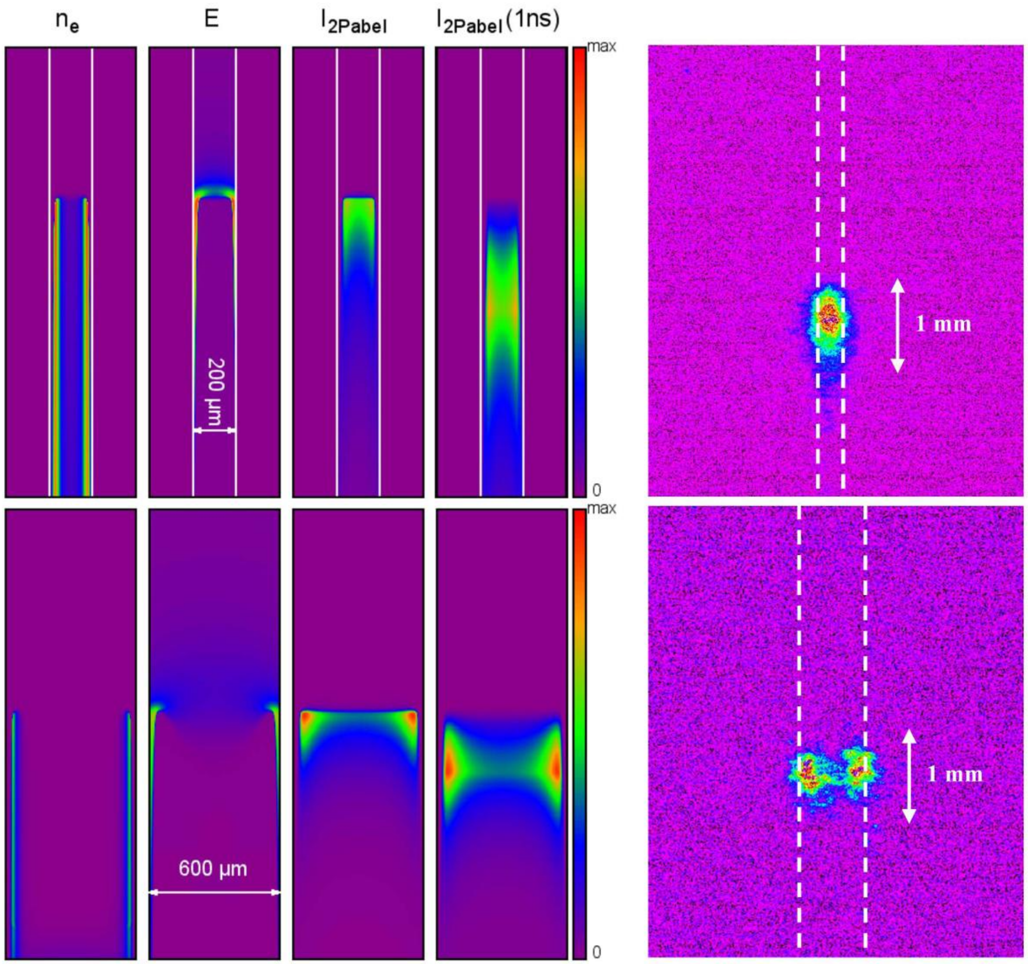
$$E_{\| gas} = E_{\| dielectric}$$

$$\nabla \cdot \vec{E} = \rho_v$$



- **Electric potential is expelled from high ε materials (solids) into low ε materials (gas).**
- **Electric field lines are bent at surfaces producing electric field enhancement.**
- **Surface charging produces parallel components of electric field.**
- **Result is *surface hugging ionization waves***

TIGHTLY BOUNDED APP STREAMERS



- **APPs also interact with their boundaries**
- **An extreme case is capillary discharges, where the channel width of the channel is commensurate with the streamer.**
- **Streamers often hug the boundaries as surface discharges.**

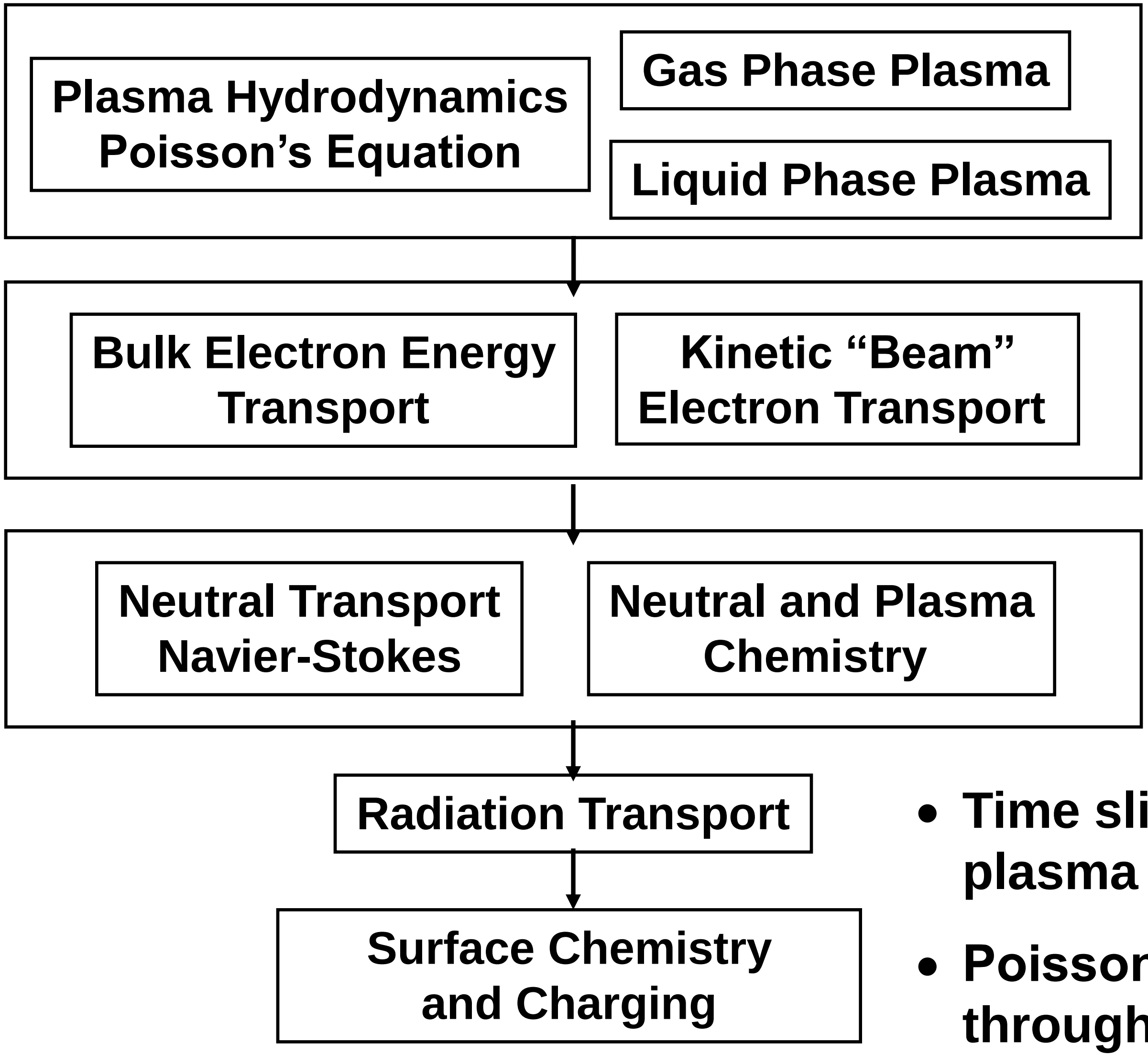
Propagation of an Air Discharge at Atmospheric Pressure in a Capillary Glass Tube

Jaroslav Jánský, Pierre Le Delliou, Fabien Tholin, Zdeněk Bonaventura, Pierre Tardiveau, Anne Bourdon, and Stéphane Pasquiers

IEEE TRANSACTIONS ON PLASMA SCIENCE

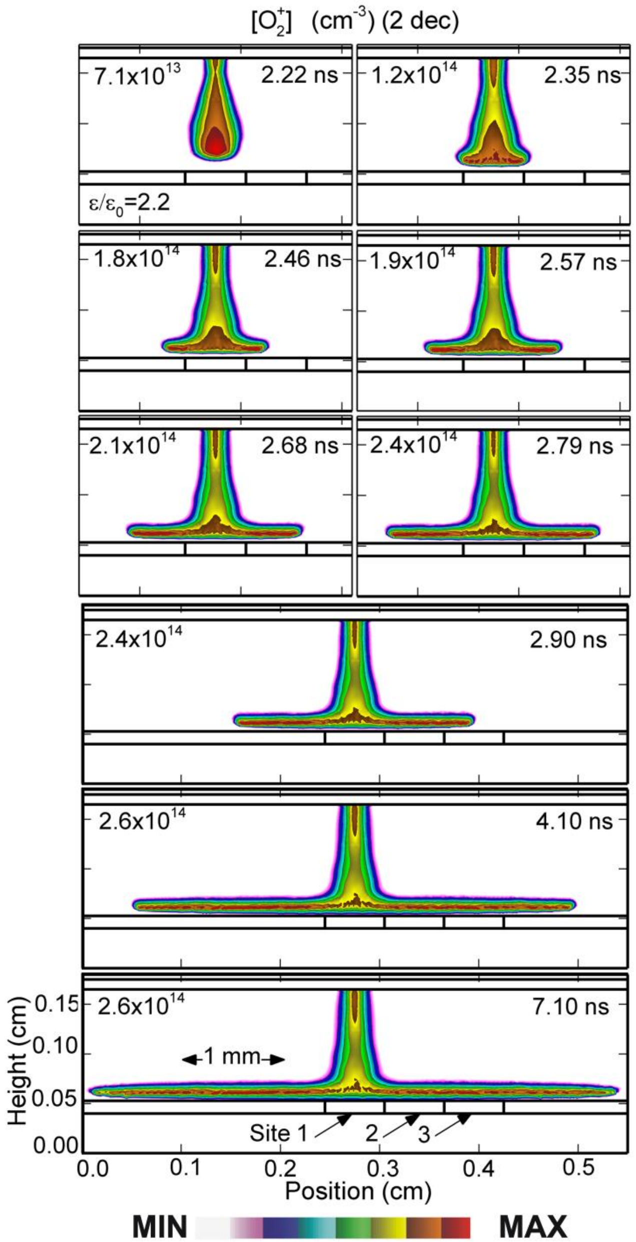
DESCRIPTION OF MODEL – AND TRANSITION TO SIW

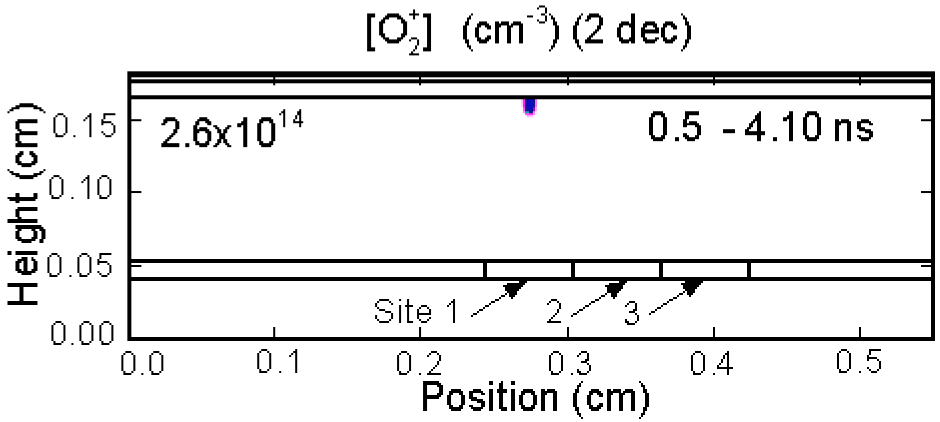
MODEL: *nonPDPSIM*



- **2-D unstructured mesh**
- **Liquid computationally treated identically to gas phase albeit different reactions.**
- **Time slicing algorithms between plasma and fluid timescales.**
- **Poisson's equation is solved throughout the computational domain including liquid.**

STREAMER DYNAMICS: OXYGEN IONS





- Electron and ion densities in the streamer channel approach 10^{15} cm^{-3}.
- Upon intersection with dielectric, the streamer charges and spreads along the surface. A wave progates outward
- Positive corona, $N_2/O_2/H_2O$ = 79.5/19.5/1.0, 1 atm, $\varepsilon_{r1} = \varepsilon_{r2} = 2.2$
- N. Babaeva, PSST 20, 035017 (2011)

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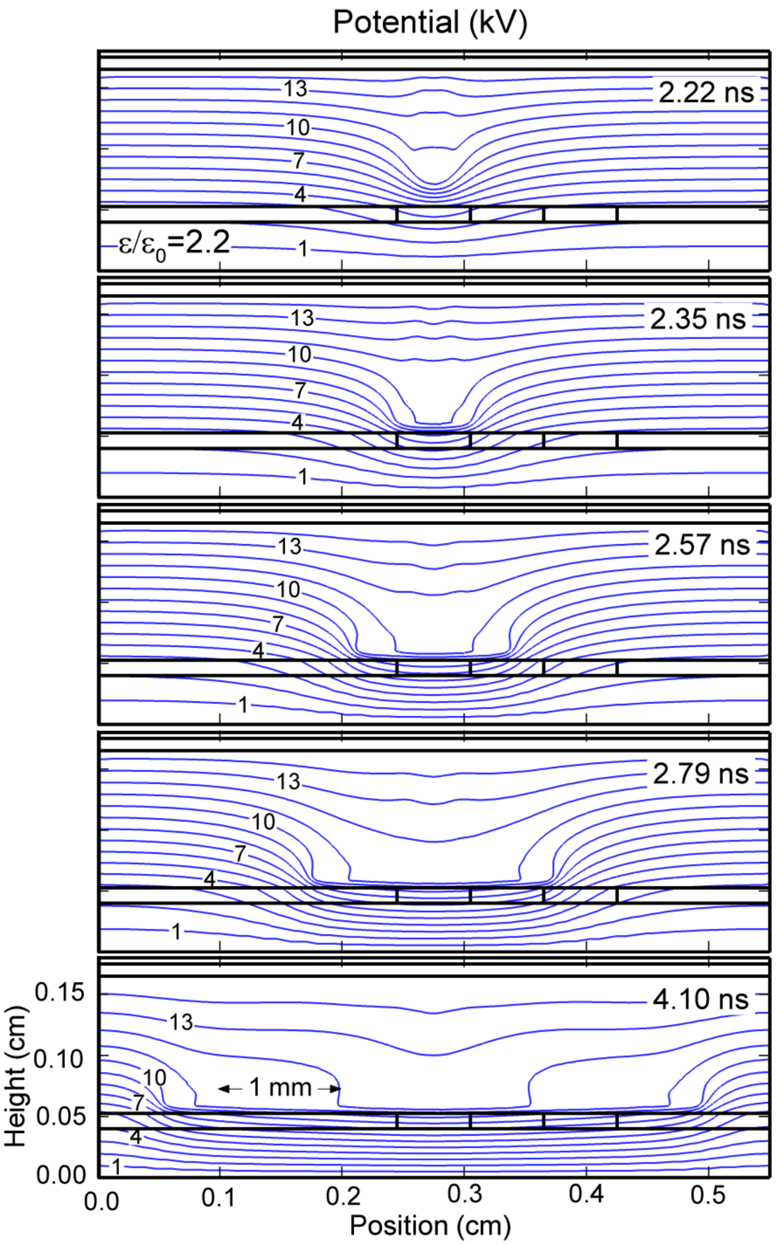
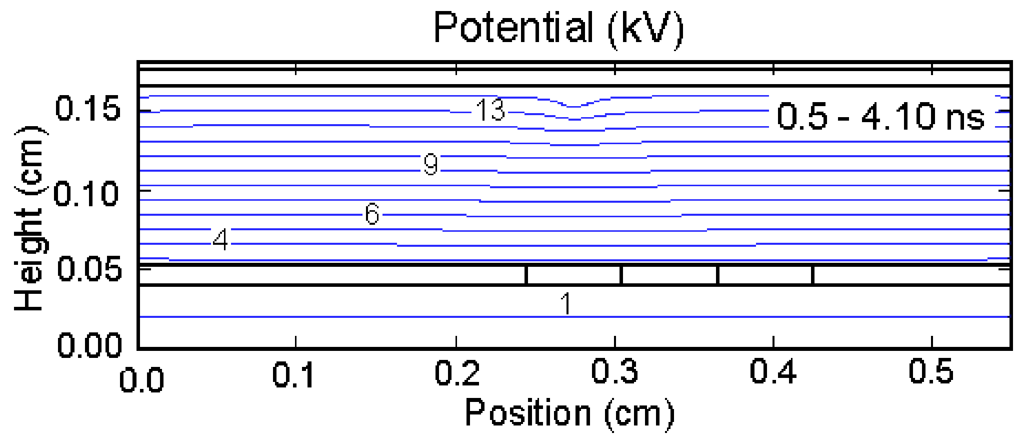
STREAMER DYNAMICS: VOLTAGE





- **As the streamer crosses the gap, voltage is shorted by the plasma column, transfering much of the applied potential to the streamer head.**
- **Upon intersection with the surface, much of this voltage is transfered first to the sheath and then to the capacitance of the dielectric.**
- **Positive corona, $N_2/O_2/H_2O$ = 79.5/19.5/1.0, 1 atm, $\varepsilon_{r1} = \varepsilon_{r2} = 2.2$**

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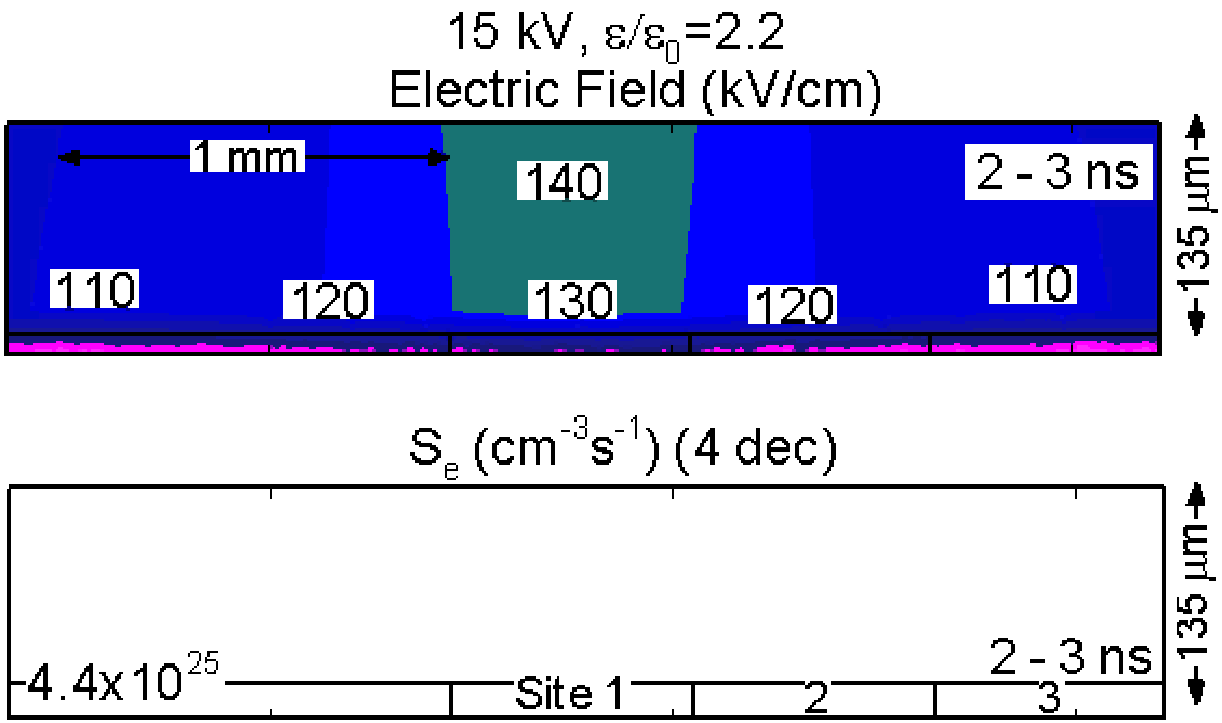
STREAMER DYNAMICS: ELECTRIC FIELD



- **In the streamer head, E-fields are 200 kV/cm when the streamer is far from surfaces.**
- **Upon intersection of the streamer with the surface, the electric fields in the resulting sheath can exceed 500-800 kV/cm.**
- **800 kV/cm x λ (0.5 μm) = 40 eV**

- **Spreading of streamer on surface produces a wave of sheath electric field away from point of intersection with E-fields of 300-500 kV/cm.**
- **Fields are radially outward at first, then oriented towards surface.**
- **Positive corona, $N_2/O_2/H_2O$ = 79.5/19.5/1.0, 1 atm, $\varepsilon_{r1} = \varepsilon_{r2} = 2.2$.**

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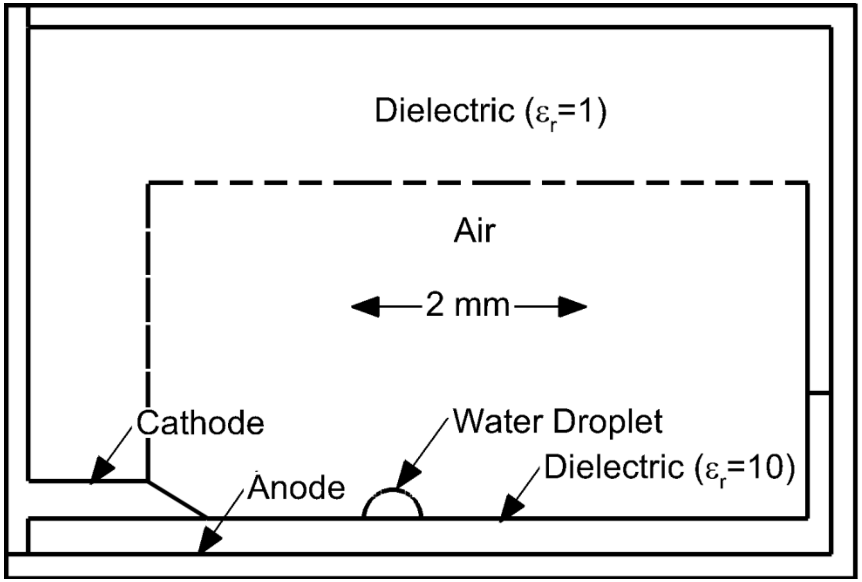
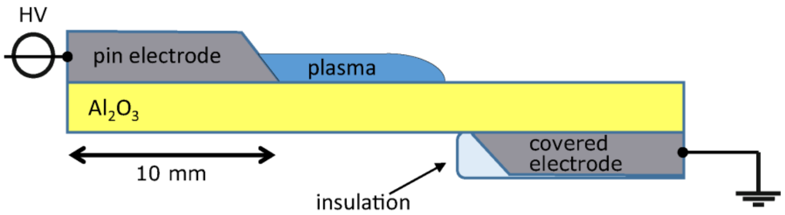
DELIBERATE SURFACE DISCHARGES OVER COMPLEX SURFACES

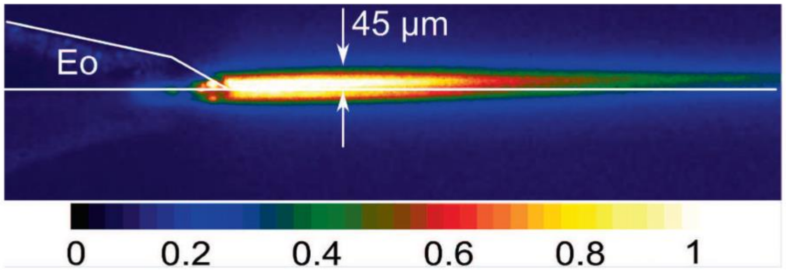
APP SURFACE DISCHARGES

- **Modeling of humid air surface discharges over nonplanar surfaces and droplets on dielectric.**
- **Deliberate launching of SIWs in a coaxial geometry.**
- **Deliberately over-voltaged to maintain SIW over long distances.**
- **Droplet study motivated by virus containing aerosol contamination.**



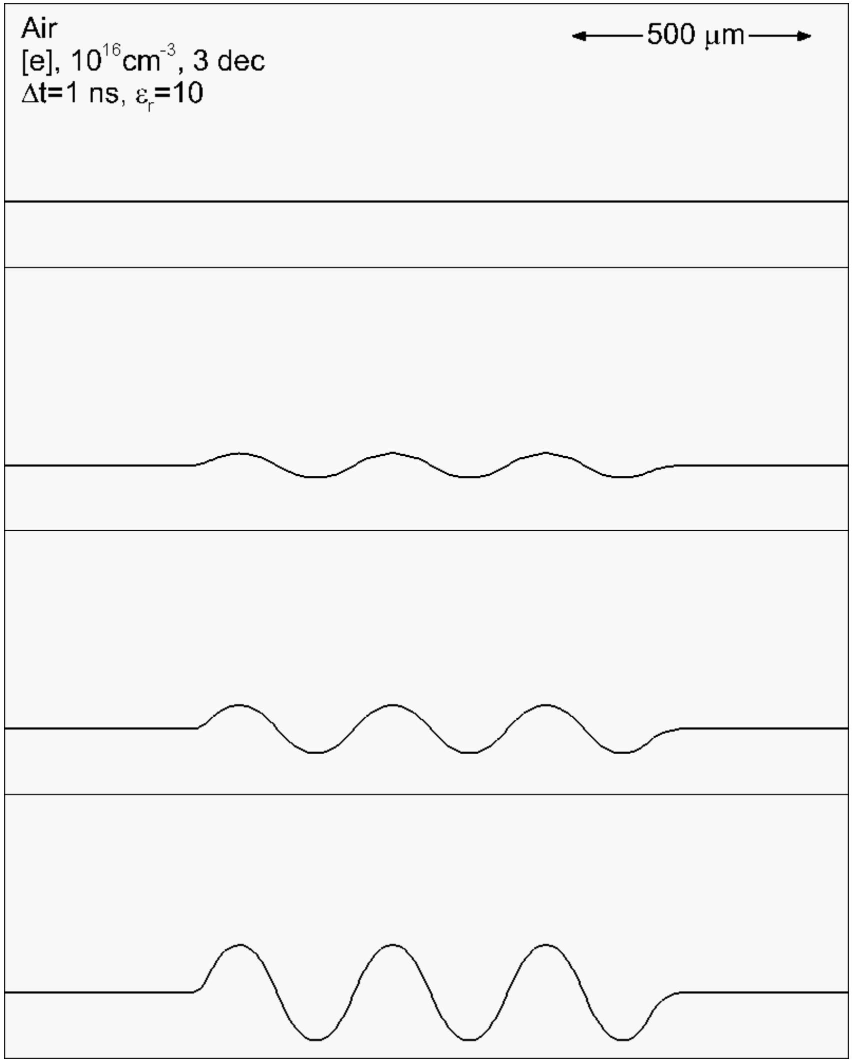
- **Surface Discharge Geometry**





- **Motivating experiments (Hoft and Brandenburg, EPJD 74, 110 (2020)).**

HUMID AIR WAVY SURFACE DISCHARGE: [e]



- Humid air, -20 kV (step), ε_r=10
- Electron density (10^{16} cm^{-3}, 3-dec)
- Surface ionization wave thickness 100 μm on flat surface.
- SIW adheres to surface for wave height < 300 μm.
- Separation of SIW from surface for deeper ridges.
- Ridges produce electric field enhancement.
- Ridges block photoelectron emission and photoionization ahead of SIW.

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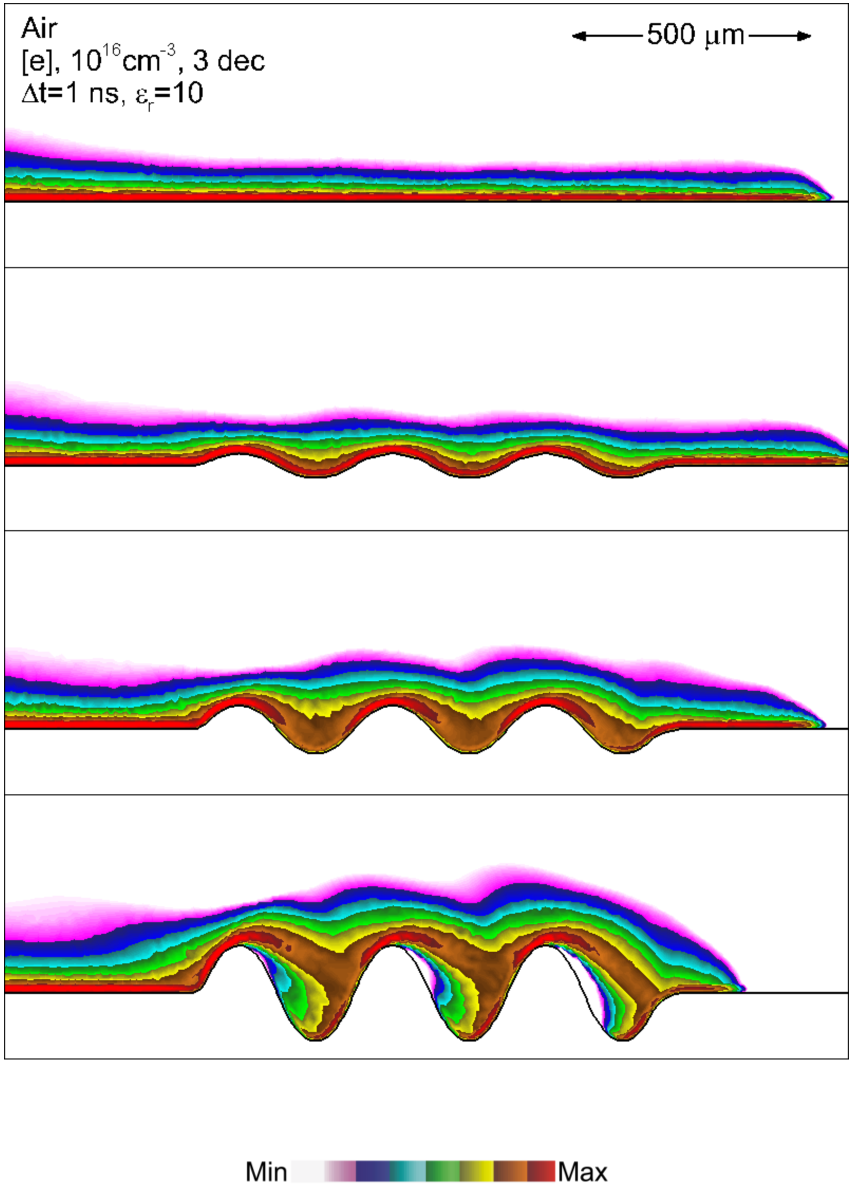
HUMID AIR WAVY SURFACE DISCHARGE: [e]



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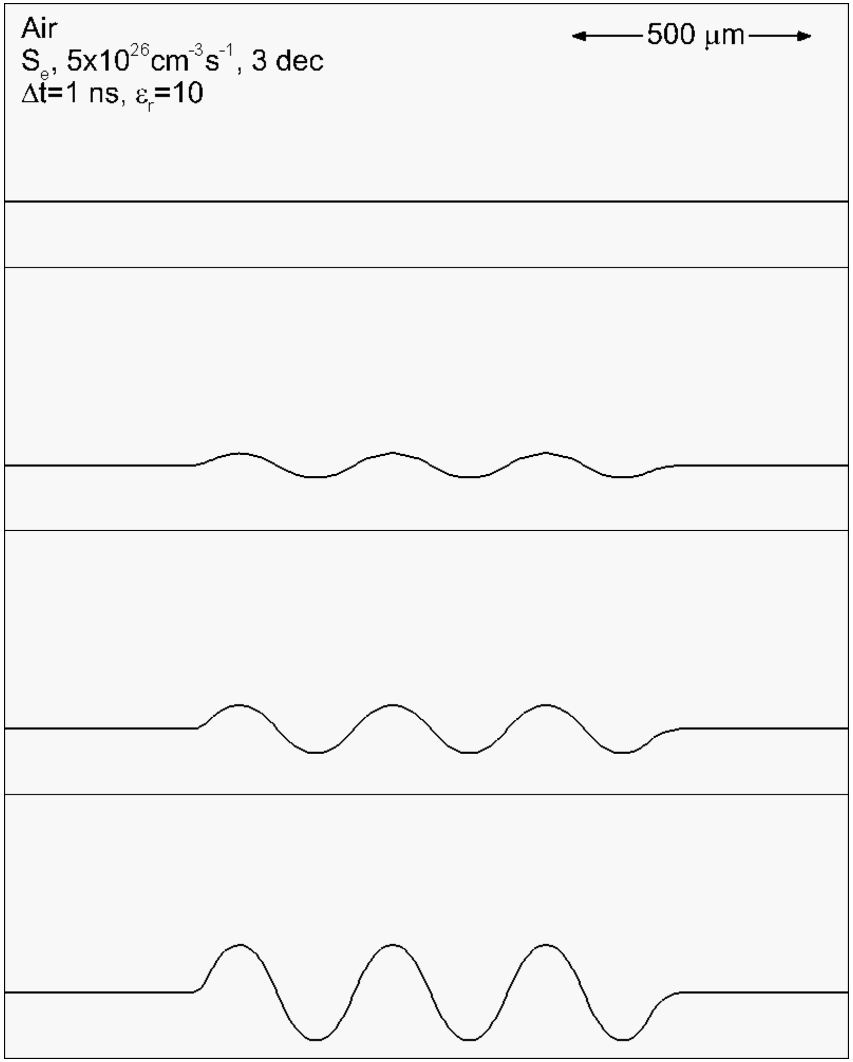
WAVY SURFACE IONIZATION SOURCE



- **Humid air, -20 kV (step), ε_r=10**
- **Ionization Rate (5x10^{26} cm^{-3}s^{-1} 3 dec)**
- **Even shallow waves produce backwards traveling surface ionization waves.**
- **Process is similar to conventional dielectric barrier discharge where surface charging launches reverse filament.**
- **Apex of wave with electric field enhancement intensifies ionization, relaunching streamer.**
- **Valleys have low electric field and slow (or quench) SIW.**

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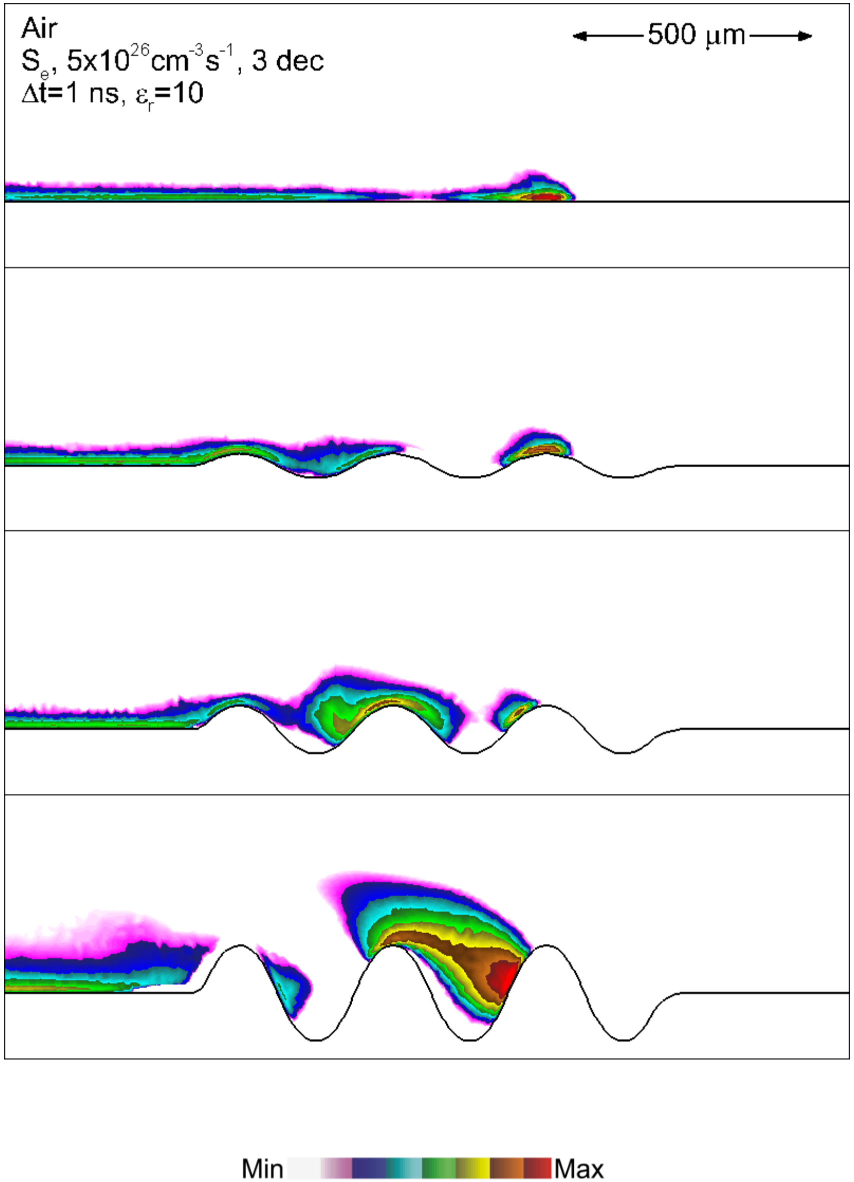
WAVY SURFACE IONIZATION SOURCE



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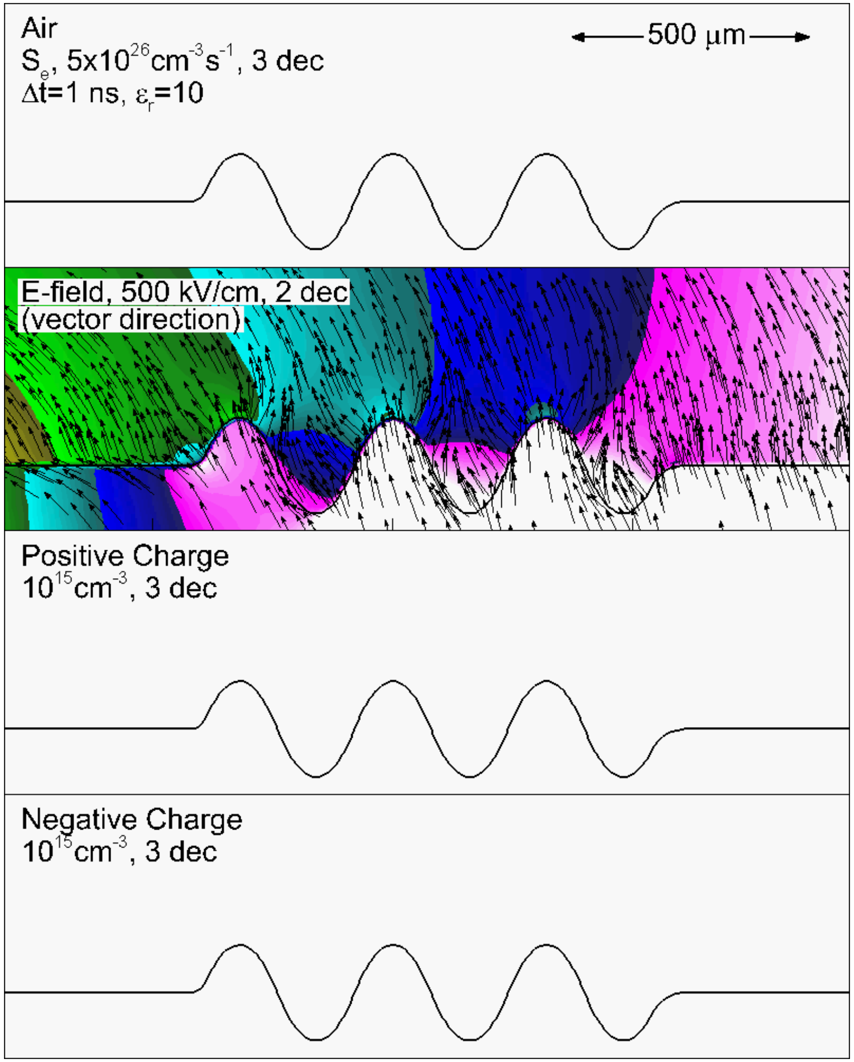
WAVY SURFACE CHARGING



- **Humid air, -20 kV (step), ε_r=10**
- **Surface charging is in large part responsible for SIW hopping and restrike.**
- **Negative SIW charges surface ahead and beneath negative, which largerly shields voltage.**
- **Positive sheath forms directly above surface.**
- **Shielded voltage facilitates field enhancement at next peak, and provides needed polarity for positive restrike.**

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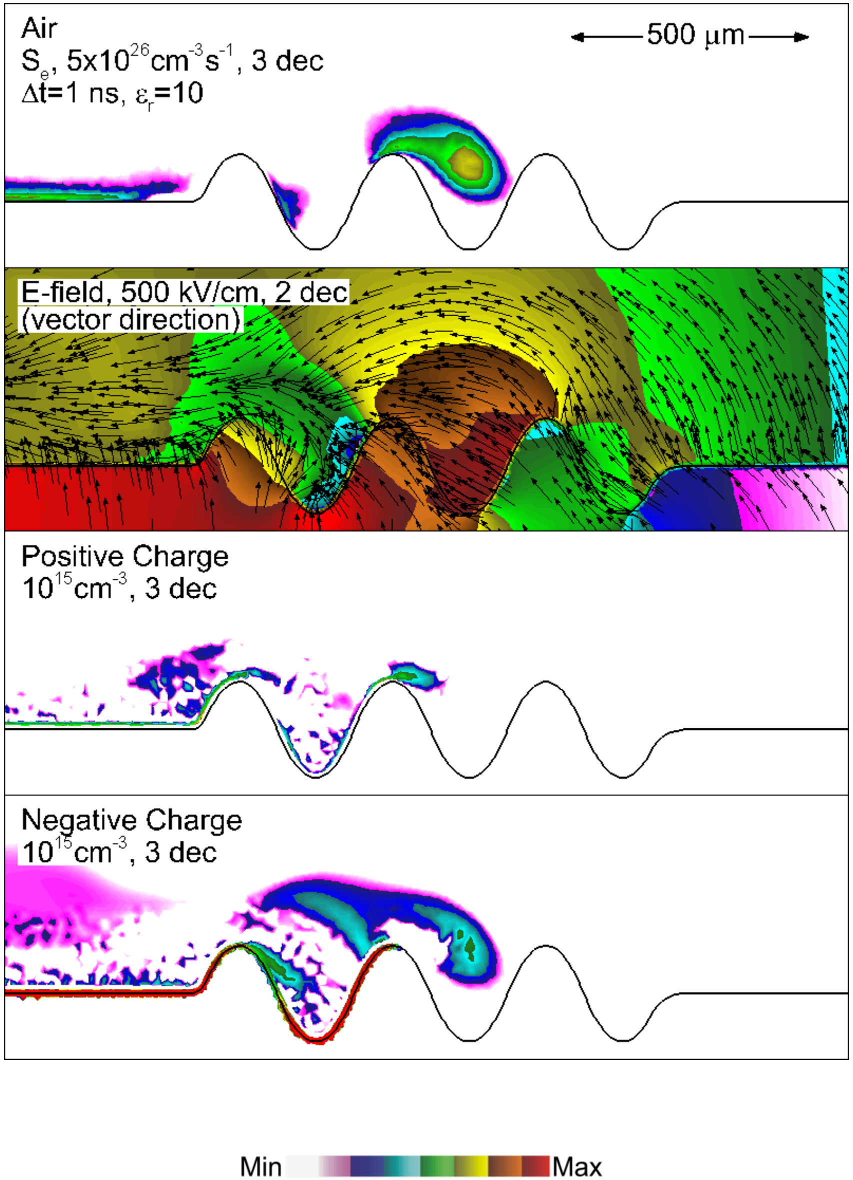
WAVY SURFACE CHARGING



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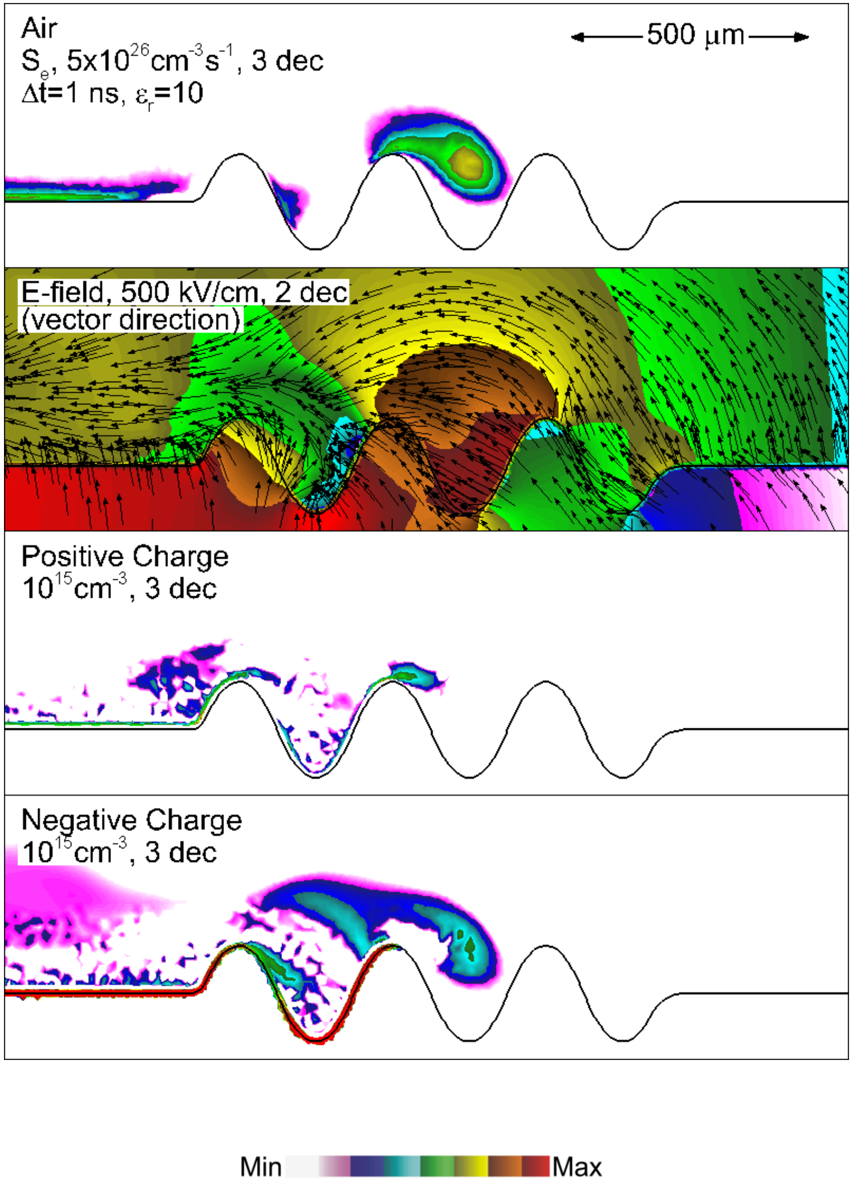

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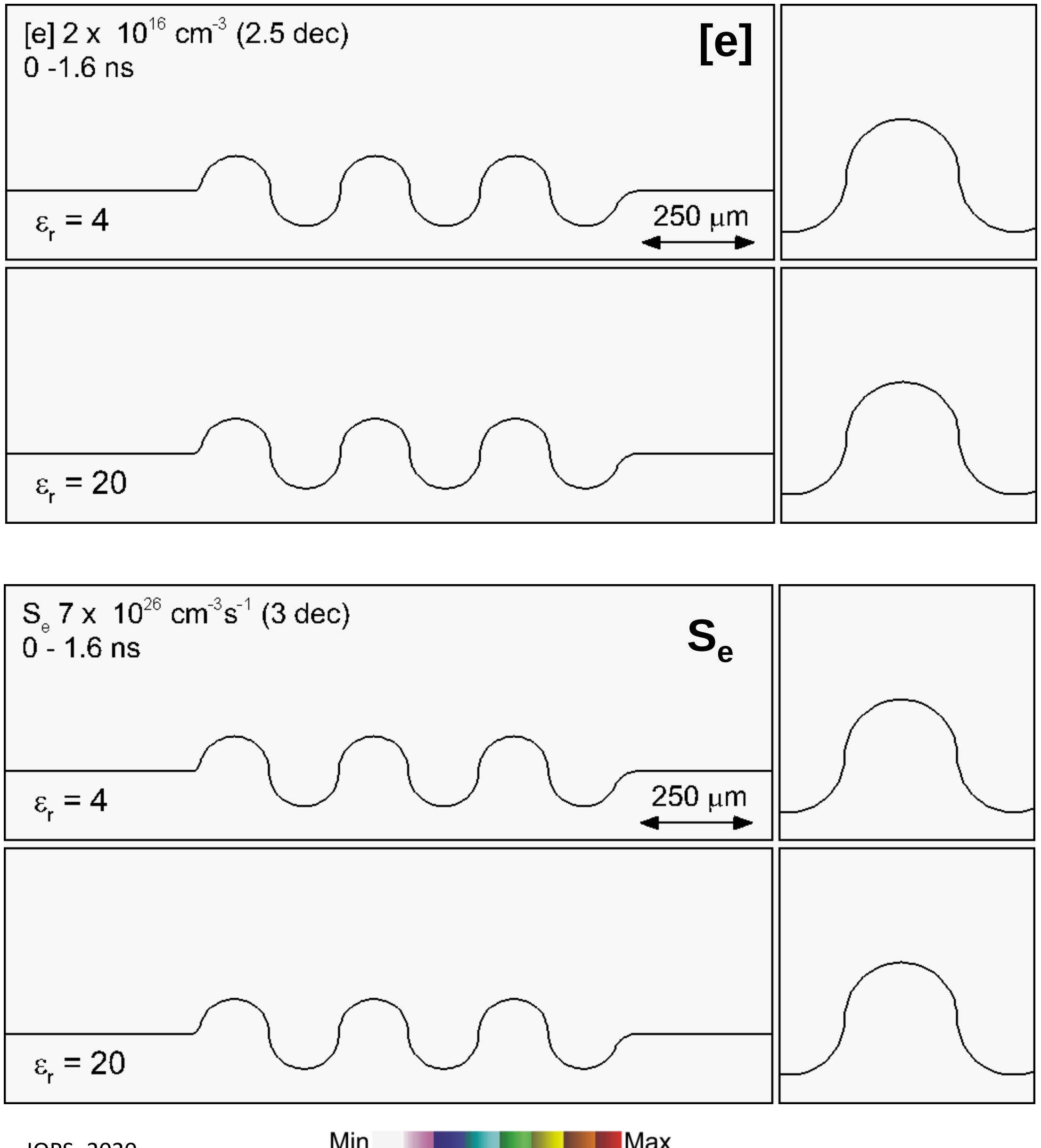
DIELECTRIC CONSTANT OF WAVY SURFACE



- **Humid air, -20 kV (step)**
- **Dielectric constant (ε_r= 4, 20) should affect propagation of SIW by:**
- **Capacitance is proportional to ε_r - larger capacitance, larger RC constant, slower propagation.**
- **Larger ε_r , more electric field enhancement at apex, producing larger ionization rates.**
- **With large over-voltage, effects are detectable, but not larger.**

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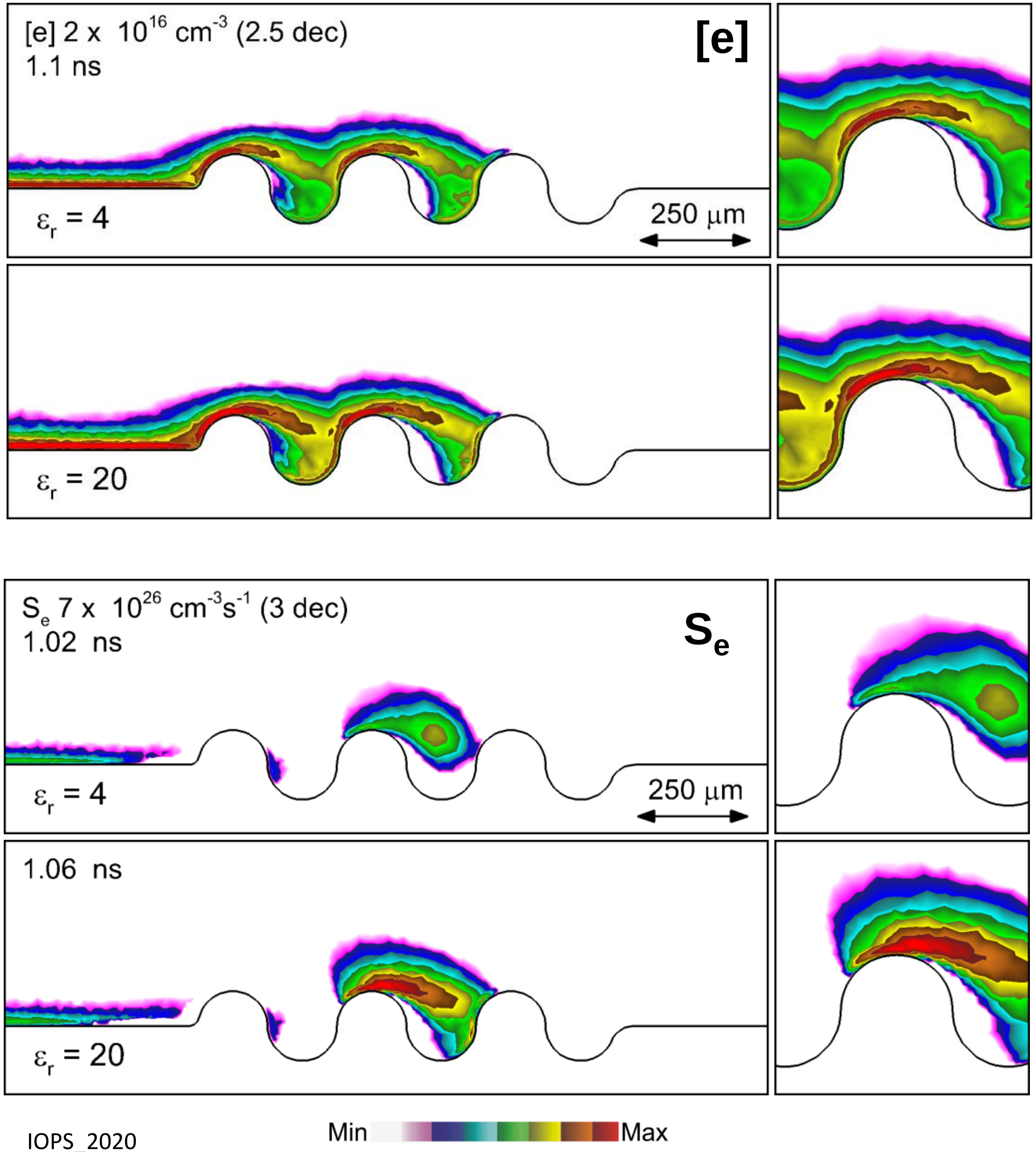
DIELECTRIC CONSTANT OF WAVY SURFACE



- **Humid air, -20 kV (step)**
- **Dielectric constant (ε_r= 4, 20) should affect propagation of SIW by:**
- **Capacitance is proportional to ε_r - larger capacitance, larger RC constant, slower propagation.**
- **Larger ε_r , more electric field enhancement at apex, producing larger ionization rates.**
- **With large over-voltage, effects are detectable, but not larger.**

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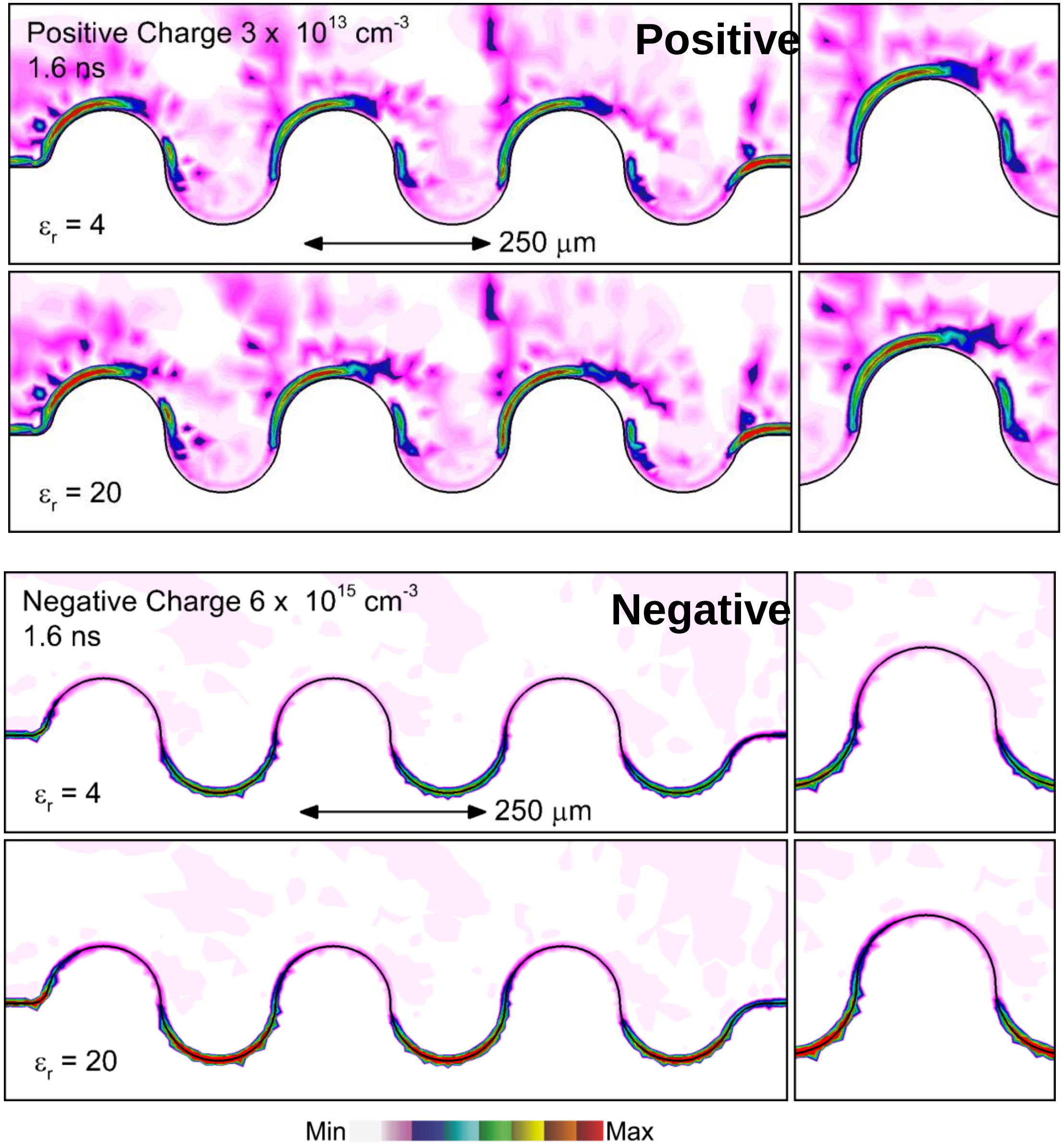
ε_r OF WAVY SURFACE: CHARGING



- **Humid air, -20 kV, ε_r= 4, 20**
- **Surface capacitance determines rate of charging, but not final state.**
- **Once surface is charged and potential established, gas phase does not see difference in ε_r (E-field enhancement aside).**
- **Sheath (and restrike remnants) not terribly sensitive to ε_r.**
- **Negative surface charging scales with ε_r to charge capacitance.**
- **Much less sensitive than packed bed reactor.**

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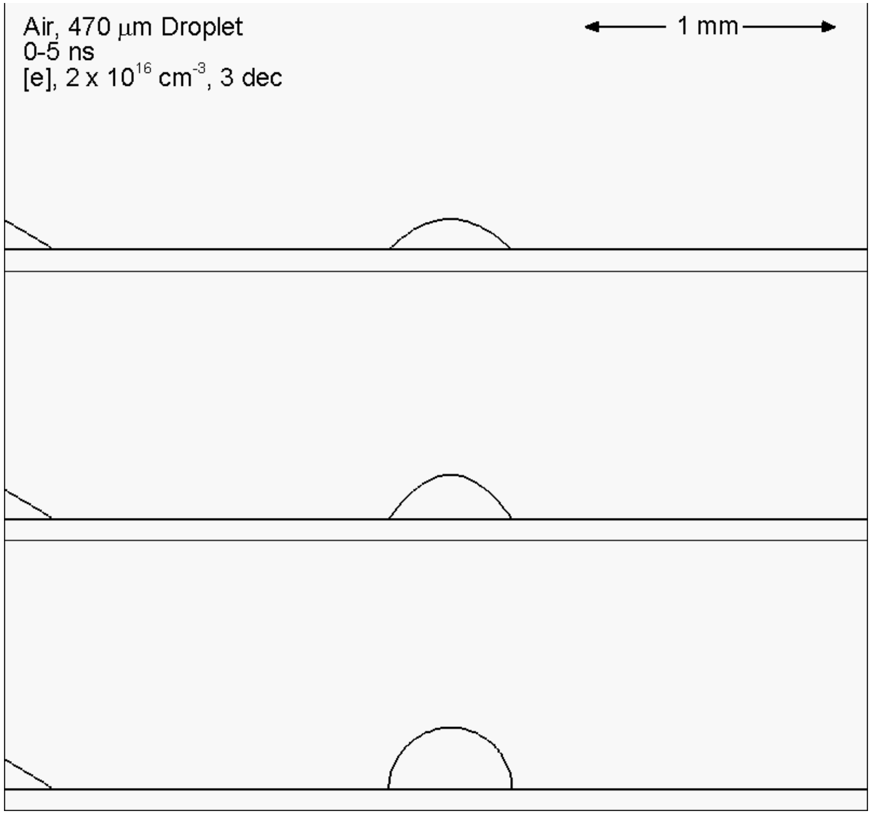
WATER DROPLET ON DIELECTRIC: [e]

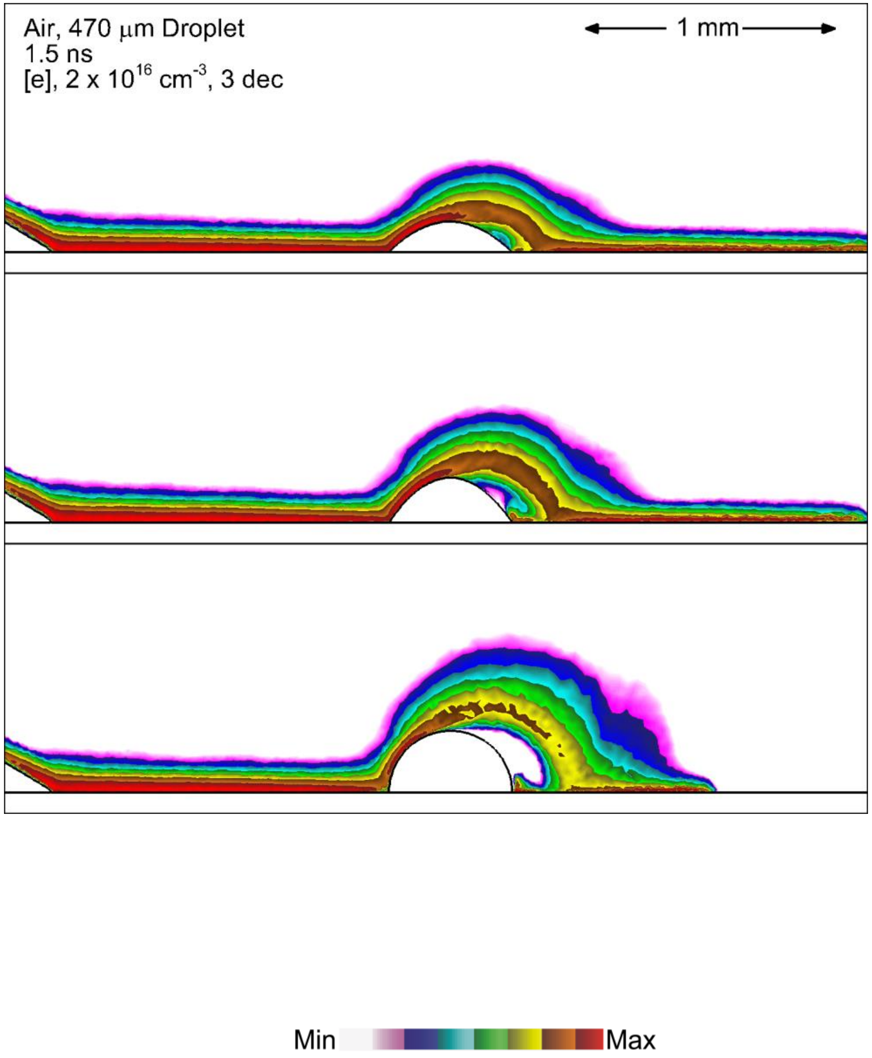


- **Humid air, -24 kV (2.5 ns), ε_r=10, electron density (10^{16} cm^{-3}, 3 dec)**
- **Motivated by plasma deactivation of virus containing aspirated aersols landing on surfaces.**
- **450 μm diameter water droplet with varying contact angle reflecting different materials.**
- **Droplets behave similar to wavy surface – plasma detaches when contact angle is small.**
- **Significant electric field enhancement at plasma-water-dielectric triple point.**

Animation Slide

Min Max

WATER DROPLET ON DIELECTRIC: [e]



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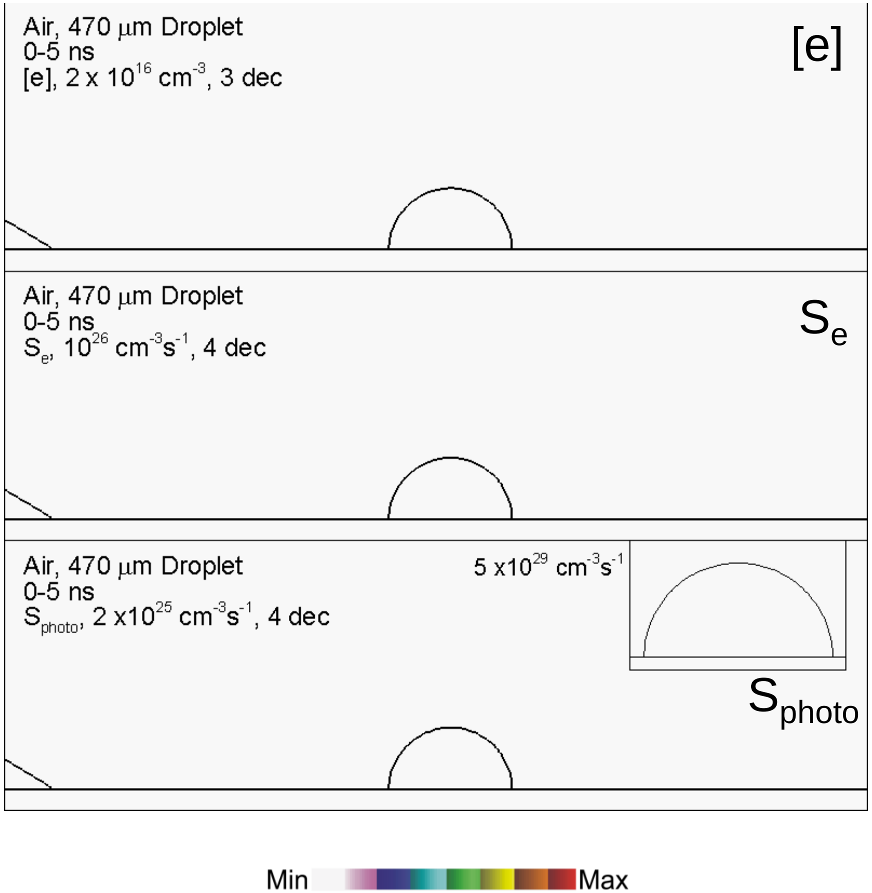
WATER DROPLET ON DIELECTRIC: IONIZATION



- **Humid air, -24 kV (2.5 ns), ε_r=10**
- **On time scale of ns pulse, water acts as a dielectric whose surface charges.**
- **Electrons immediate solvate but do not convect far from surface.**
- **Electric field enhancement at leading corner attracts SIW.**
- **Surface charging launches ionization wave over droplet.**
- **Photoionization at surface of droplet under SIW is 10^3-10^4 times more intense than in gas phase.**
- **Majority of immediate activation is VUV dominated.**

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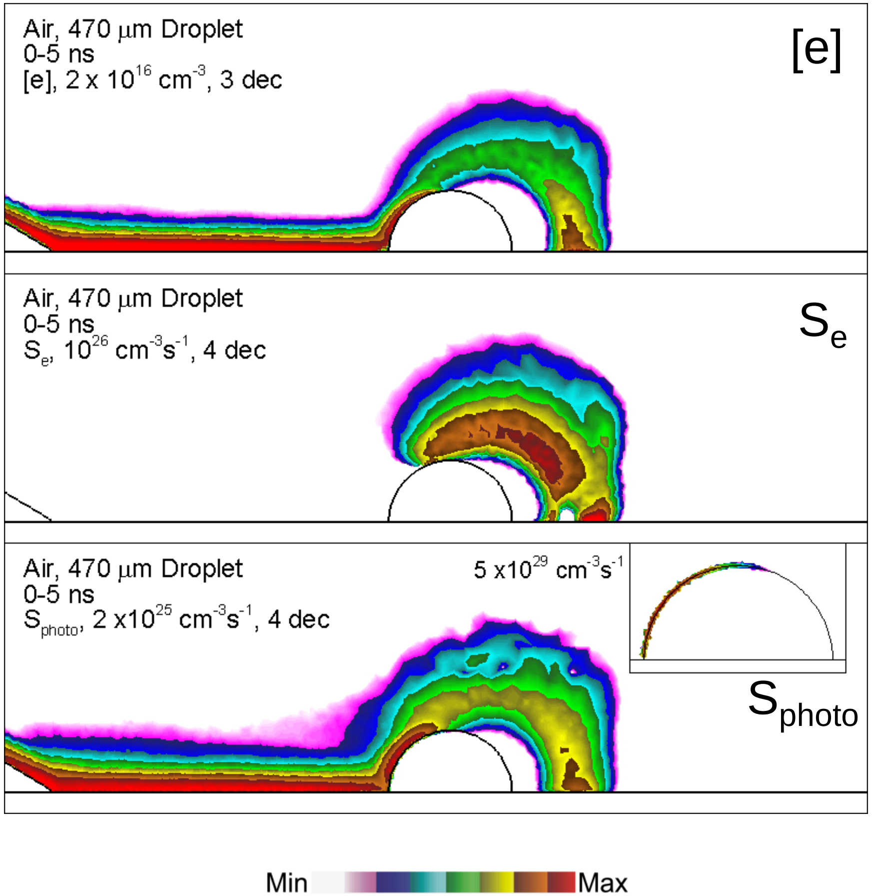
WATER DROPLET ON DIELECTRIC: IONIZATION



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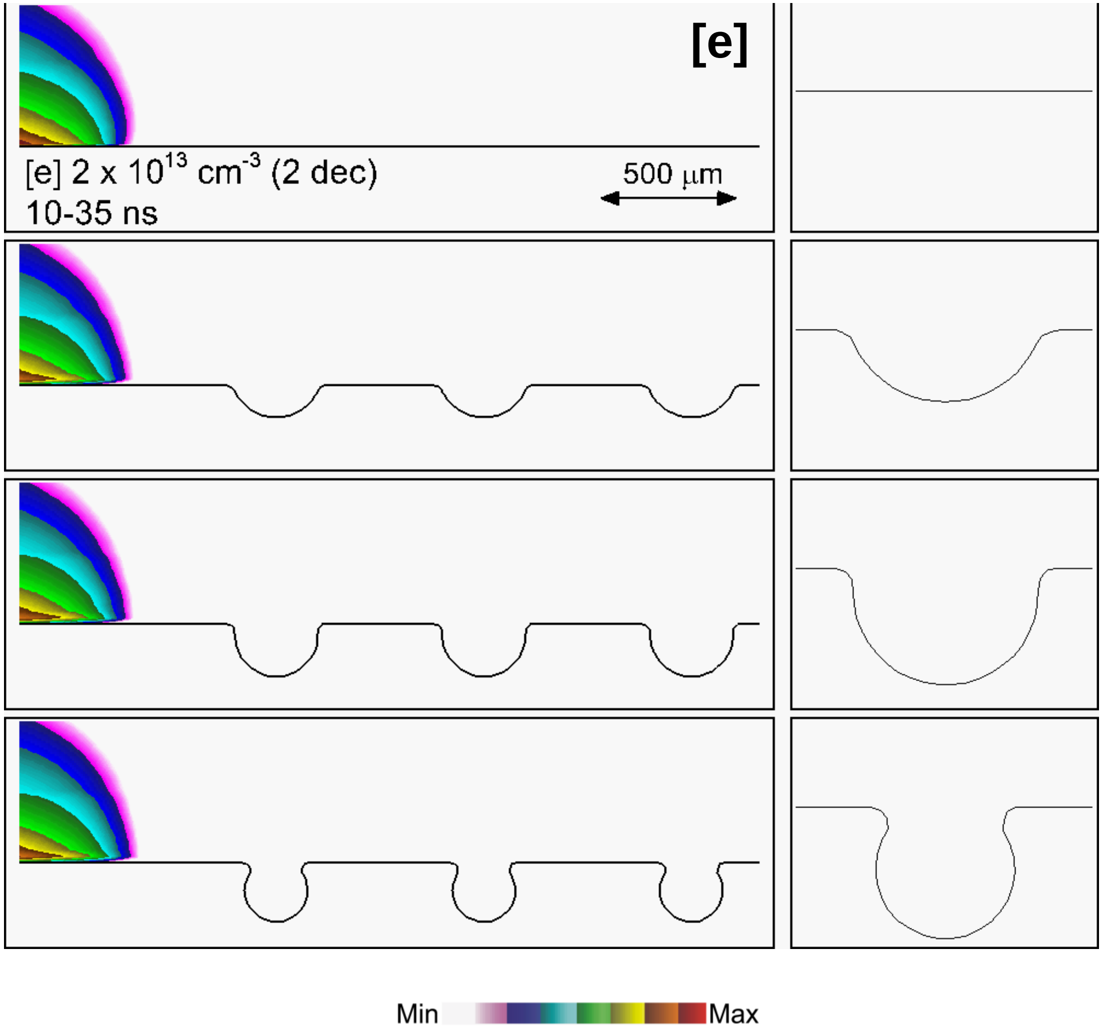
APPs ACROSS POROUS MATERIALS

- Propagation of APPs across (or onto) porous materials is senstive to the “cut” through the materials.
- Pores can be:
  - Subsurface
  - Barely opened
  - “Sliced open”.
- Pores are typically interconnected, enabling pore-to-pore propagation.
- Do “inverted” waves behave the same?
- How do plasmas get into pores?

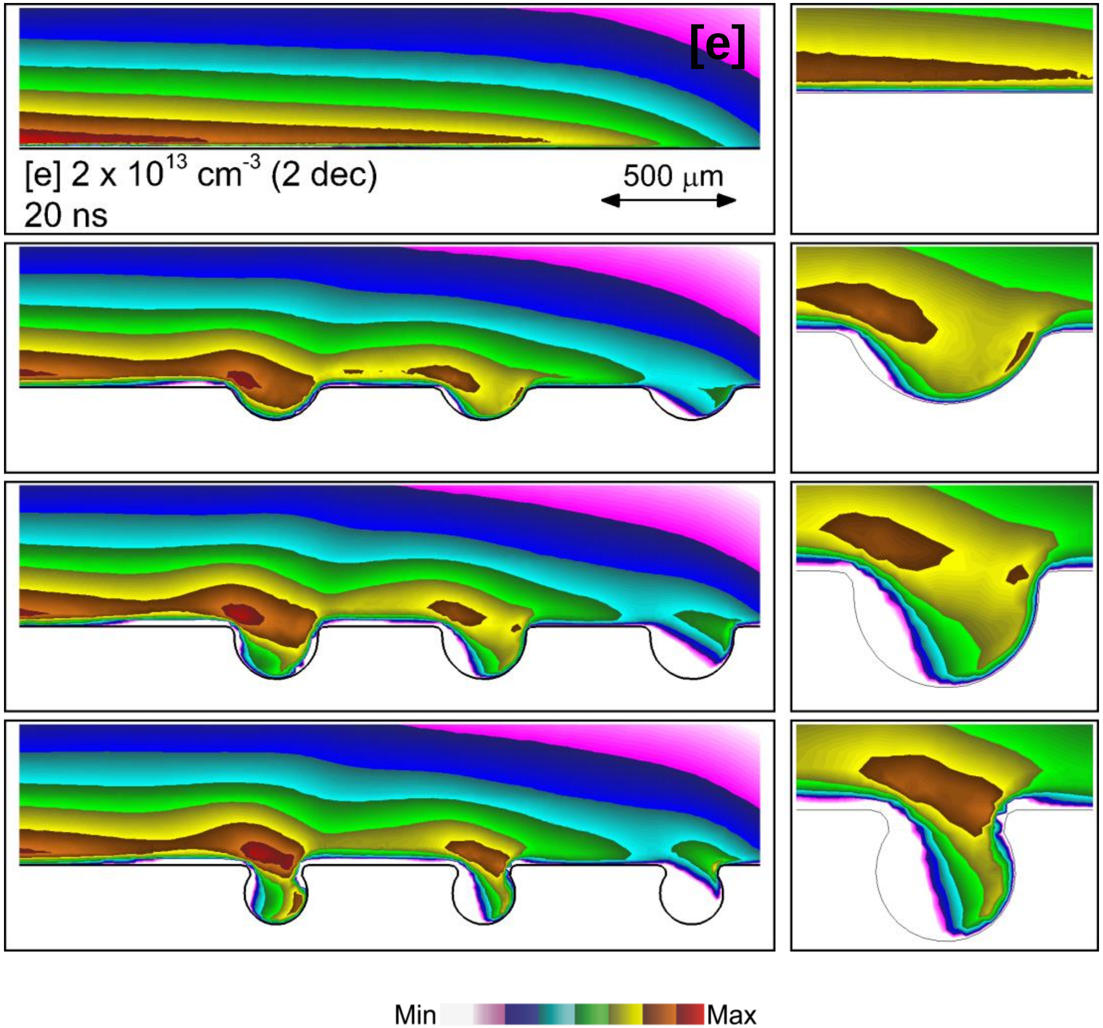
"CUT" THROUGH POROUS MATERIAL: [e]



- Humid air, -25 kV (step), ε_r=4
- Electron density
- APPs react to cut pores similarly to wavy surfaces.
- Interaction is here more dominated by electric field enhancement at edges of cut pores.
- Overshoot of pore due to E-field enhancement on opposite edge leads to unexposed surfaces.
- Charging dominates in production of restrike SIWs.

Animation Slide

"CUT" THROUGH POROUS MATERIAL: [e]



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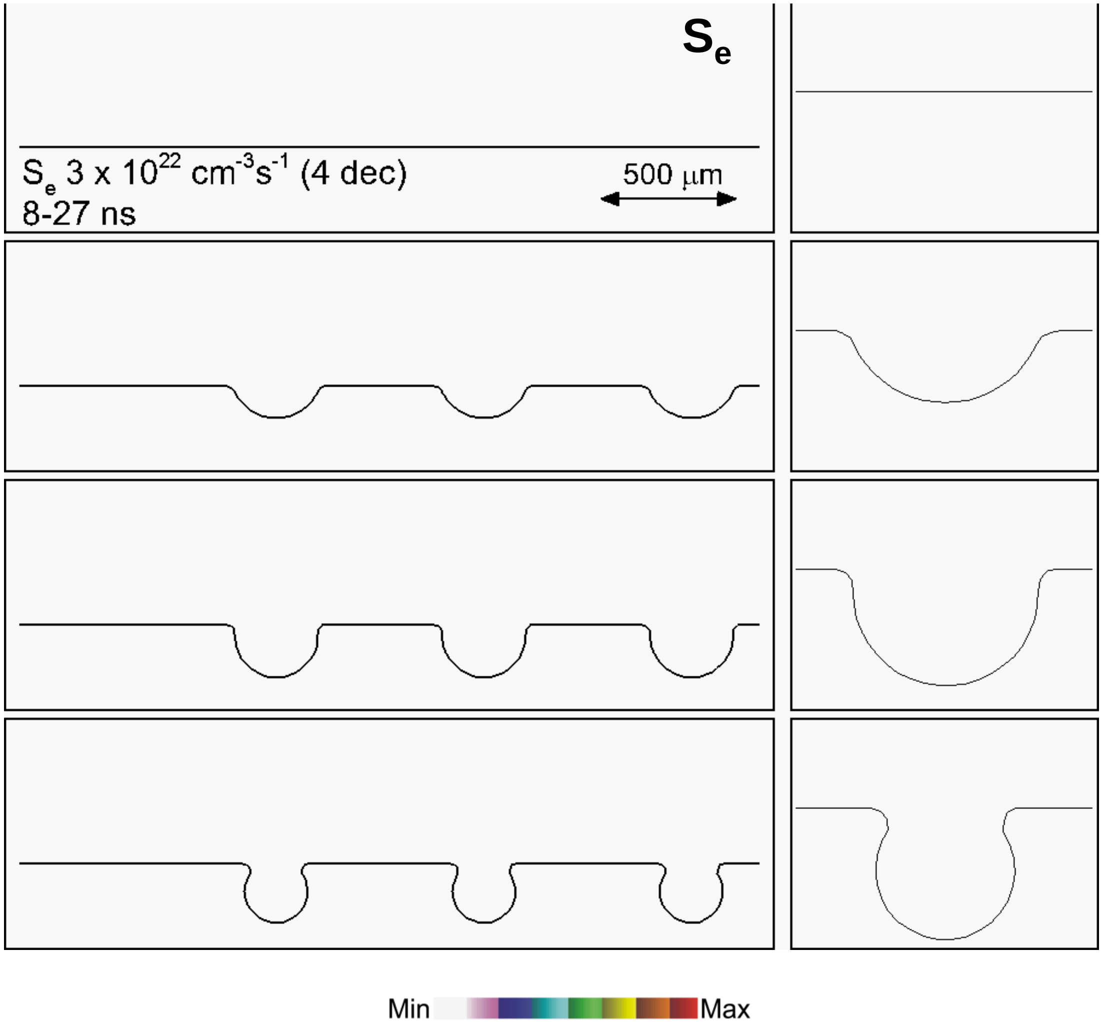
"CUT" THROUGH POROUS MATERIAL: S_e



- Humid air, -25 kV (step), ε_r=4
- Ionization source.
- Sharp edges produced electric field enhancement and have larger capacitance.
- Intense ionization at "more than half pore" edges
- Charged induced restrike SIW provides full coverage of shallow pores.
- Restrike SIW in greater than half-pores are not able to fully cover the inner surface.

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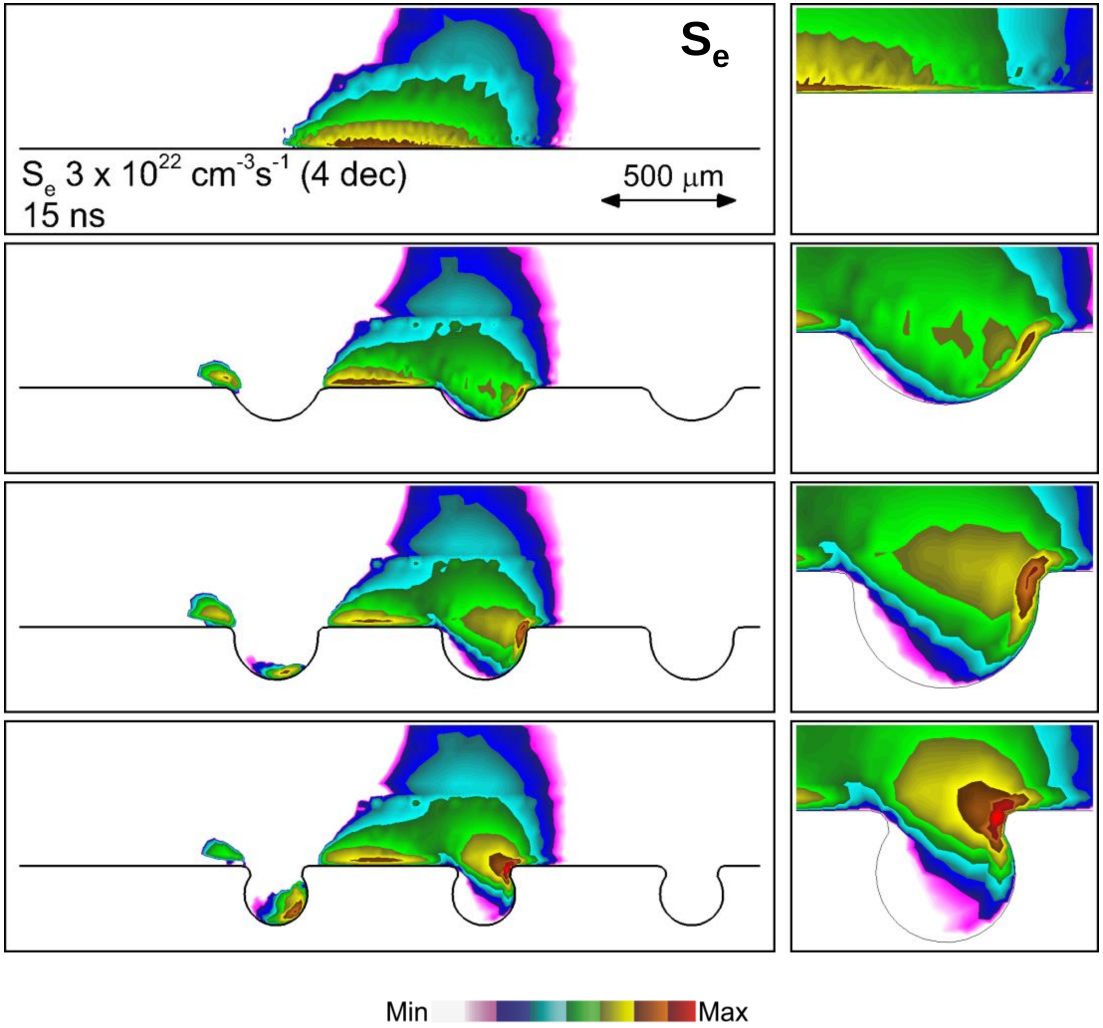
"CUT" THROUGH POROUS MATERIAL: S_e



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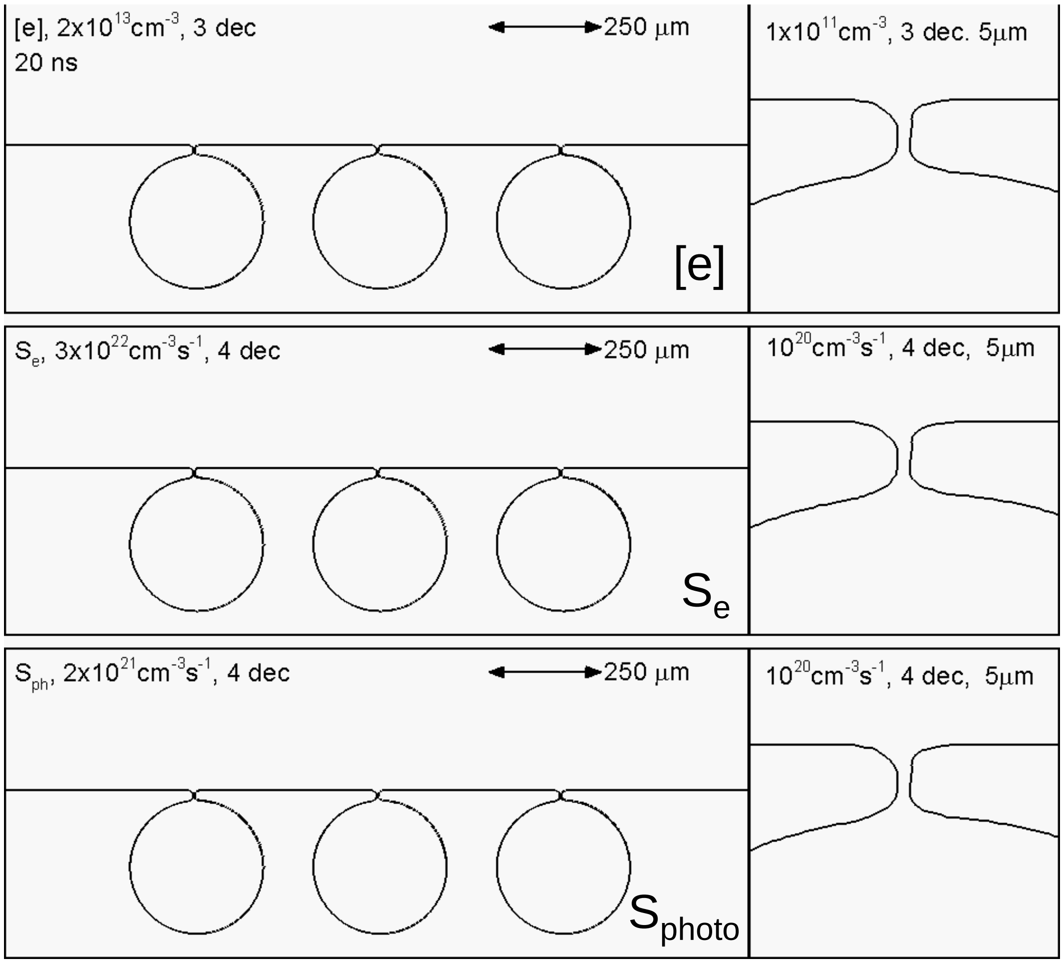
SIW OVER SUBSURFACE (OPEN) PORES



- **Humid air, -25 kV (step), ε_r=4**
- **Flat surface with subsurface pores (300 µm)**
- **5 µm opening to plasma to small to enable plasma to naturally flow (λ_D= 3-4 µm). Leakage is non-ambipolar.**
- **VUV illumination through pore opening seeds plasma.**
- **Plasma “sees” subsurface pore through lower capacitance of underlying dielectric.**
- **Electric field enhancement at opening.**

Animation Slide

Min Max

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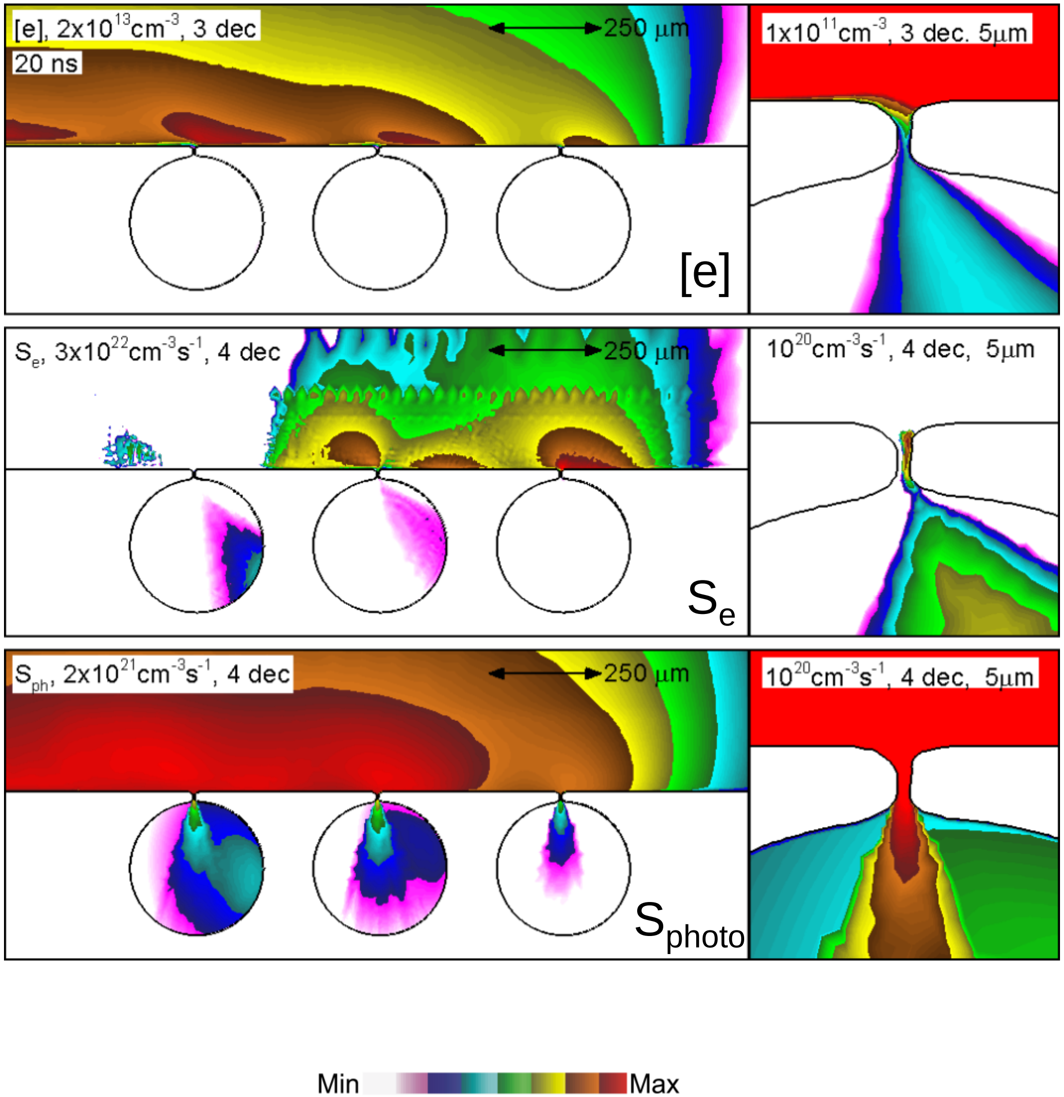
SIW OVER SUBSURFACE (OPEN) PORES



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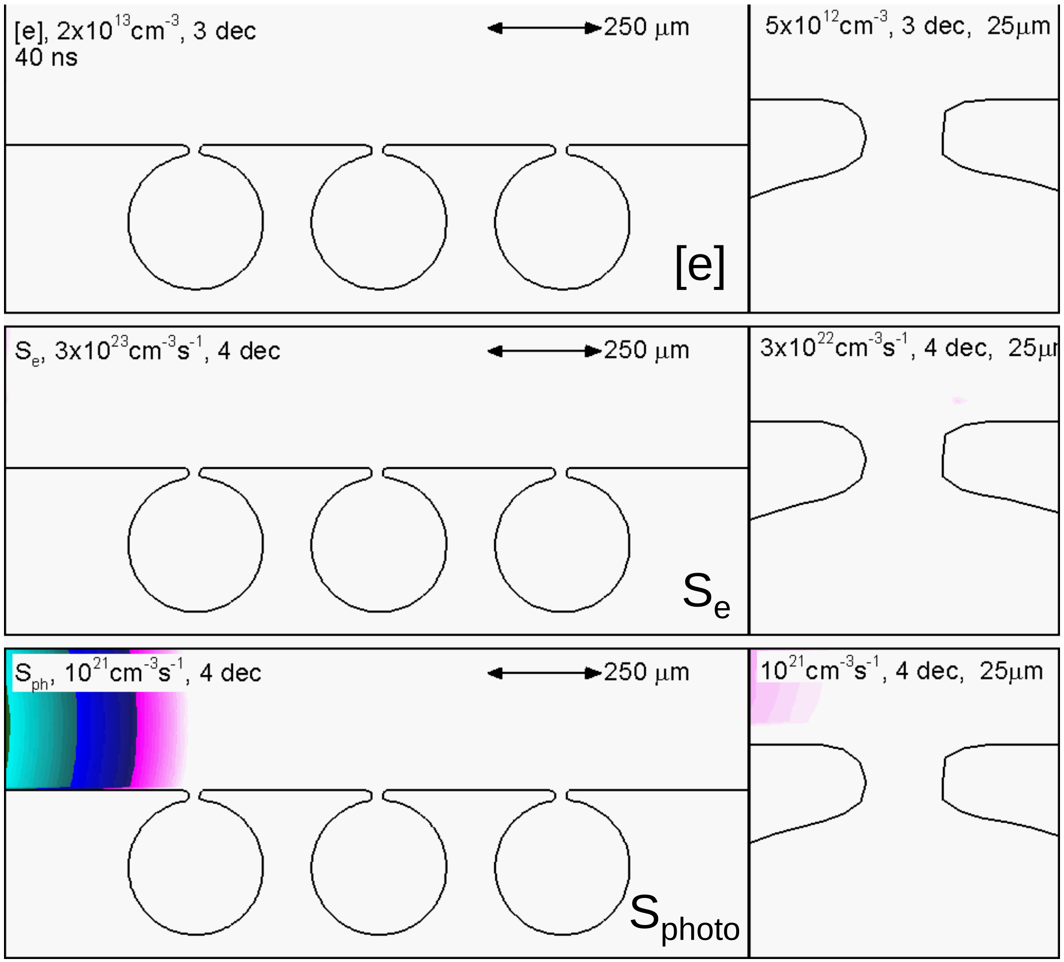
SIW OVER SUBSURFACE PORES: LARGE OPENING



- **Humid air, -25 kV (step), ε_r=4**
- **Flat surface with subsurface pores (300 μm)**
- **25 μm opening to plasma to exceeds Debye length and allows flow of plasma into pore.**
- **VUV illumination seeds plasma, though electron impact dominates ionization.**
- **Plasma "sees" subsurface pore through lower capacitance of underlying dielectric.**
- **Pores see each other through charging of surfaces.**

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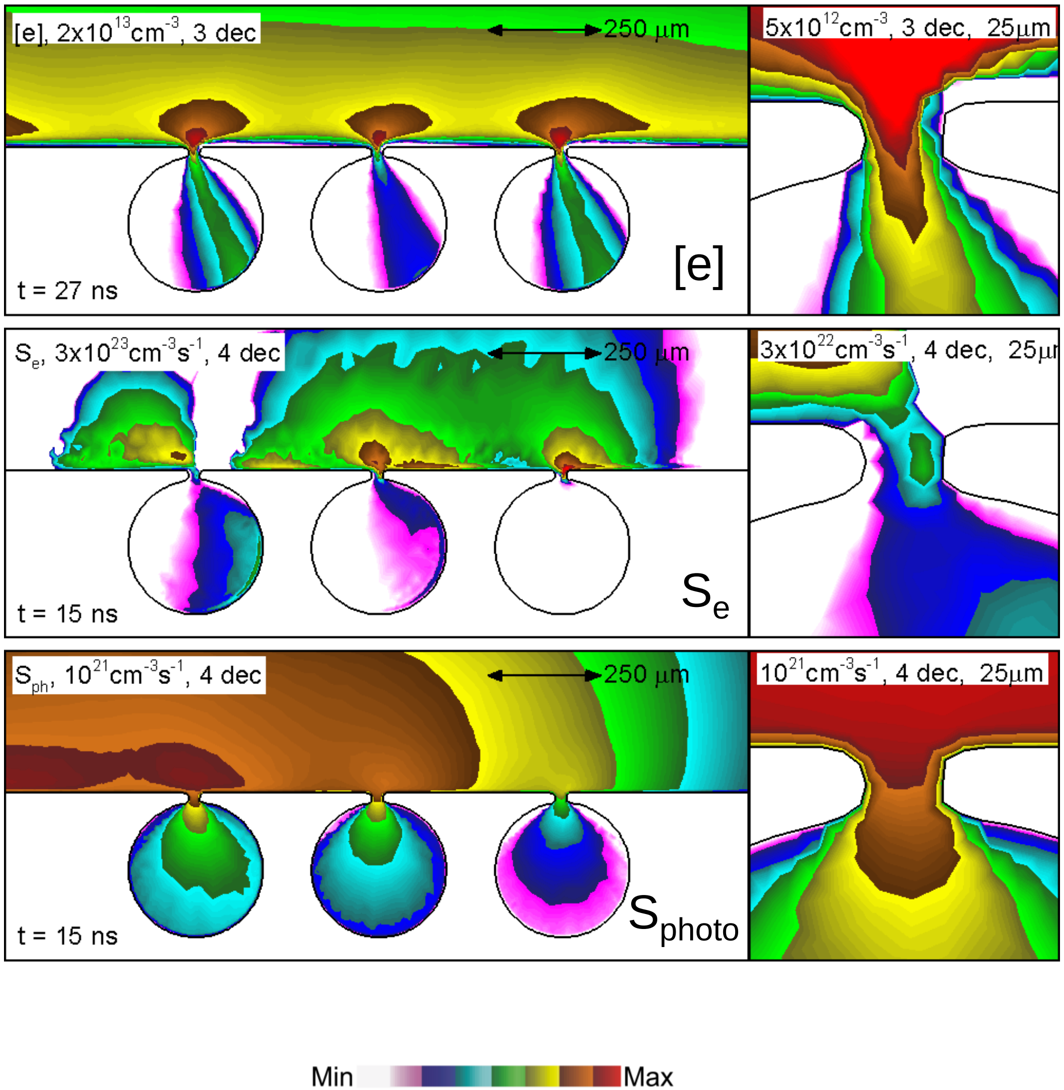
SIW OVER SUBSURFACE PORES: LARGE OPENING



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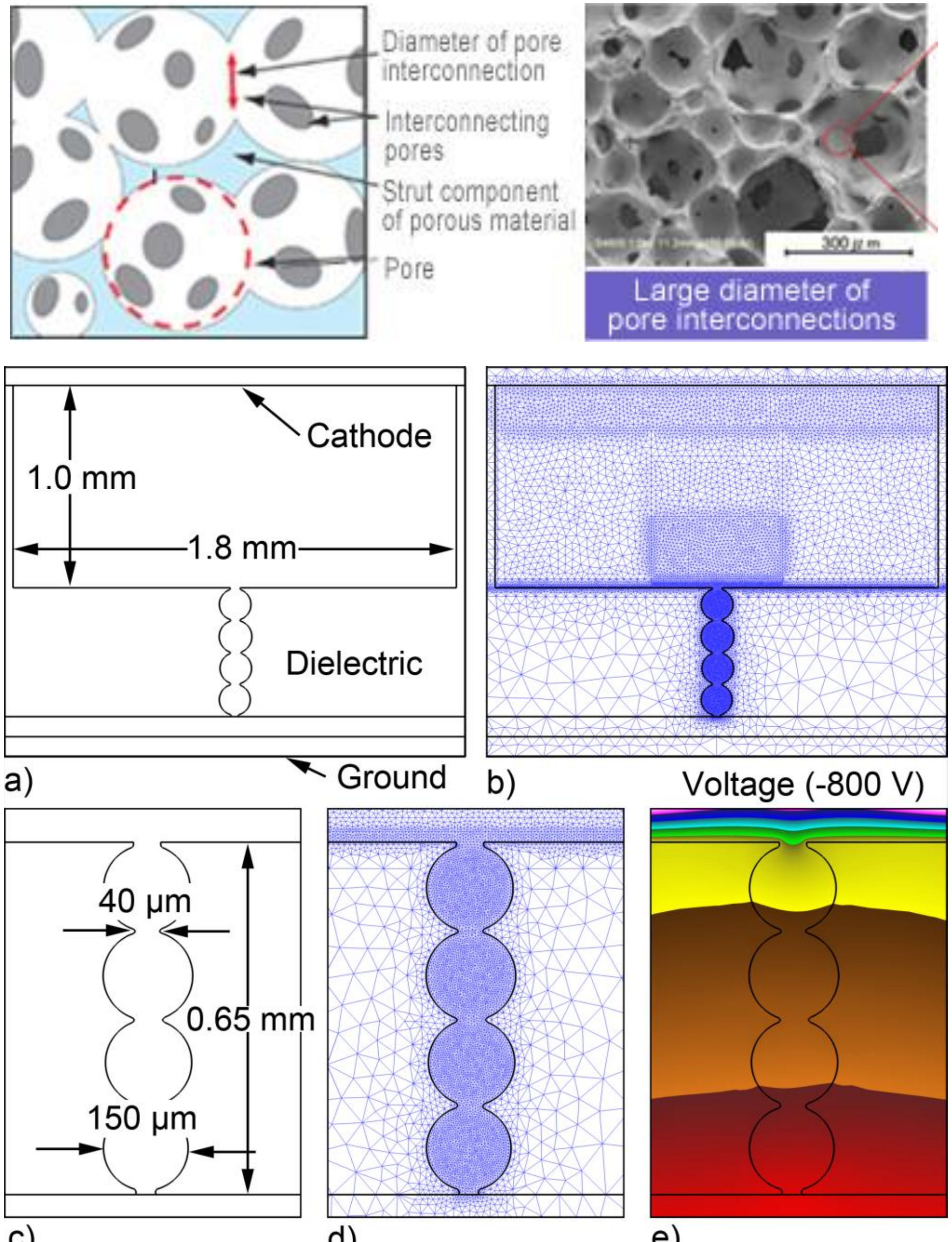
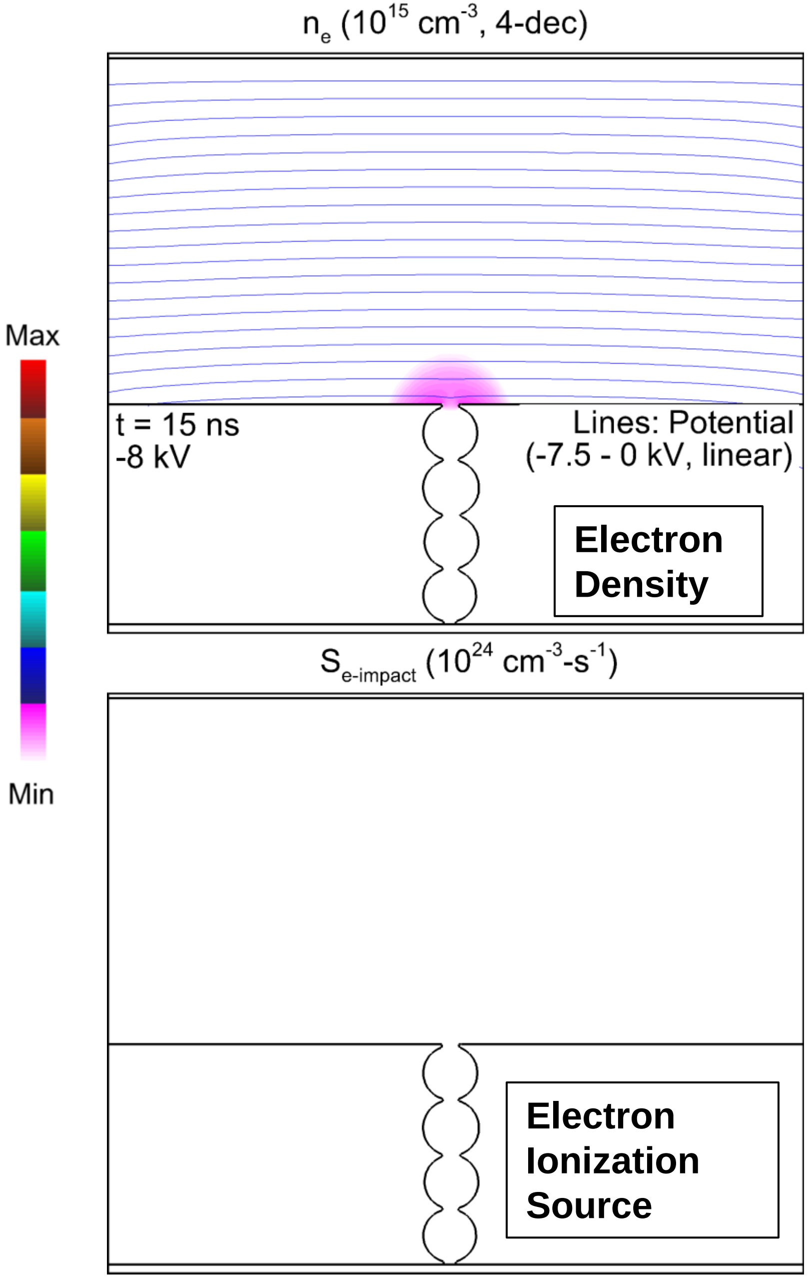
CONNECTED CHAINS OF PORES



INTERCONNECTED PORES

- Treatment of internal surface of interconnected pores of interest to tissue scaffolding.
- Examine single chain of pores.
- Dielectric barrier discharge (DBD) in humid air ($N_2/O_2/H_2O$= 78/21/1).
- 1.0 mm gas gap, 0.65 mm dielectric layer (ε_r = 61, wet bone).
- -8 kV, 15 ns, top electrode.

MICRODISCHARGE FORMATION



- **Electron density, equipotential lines, and electron impact ionization source.**
- **Townsend avalanche begins near cathode, electrons impact and charge scaffold.**
- **Restrike propagates towards cathode.**
- **Surface ionization waves develop on scaffold.**
- **Conductive plasma shorts potential in pores, producing high electric field.**
- **Discharges take place inside pores.**

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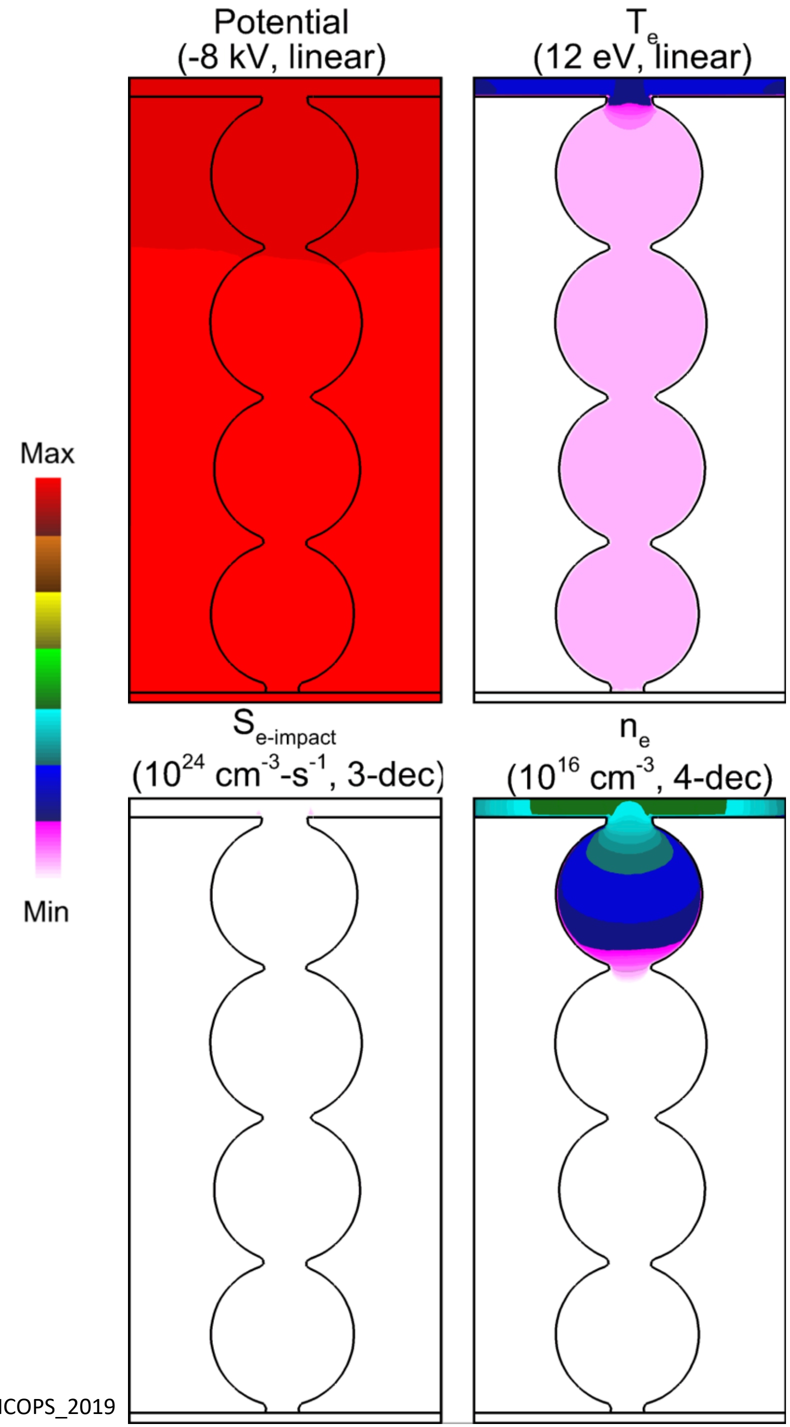
PROPAGATION INTO PORES



- **Initially, electrons enter pores following electric field.**
- **Potential drop over small distance leads to high electron temperature (peak at $T_e \approx 11.7$ eV).**
- **Microdischarges form between pore connectors.**
- **Charging of connectors leads to formation of surface ionization waves (SIWs).**

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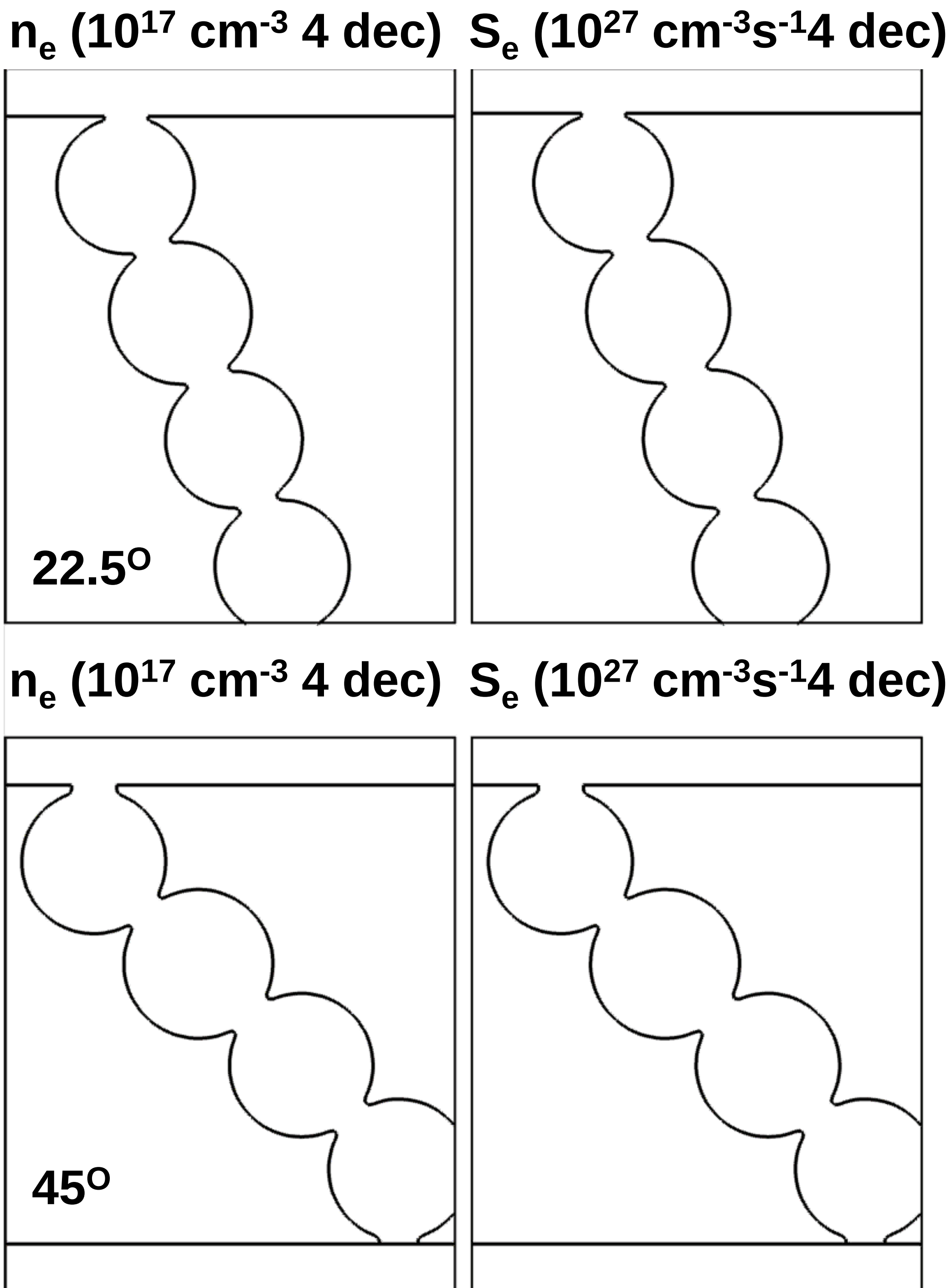
PORE ORIENTATION



- Original orientation of electric field is imprinted on IW propagation.
- Oblique pores are increasingly shadowed have IW on bottom surface.
- Restrike SIW fails to cover all surfaces.
- 1 atm, humid air, -8 kV , 5 ns

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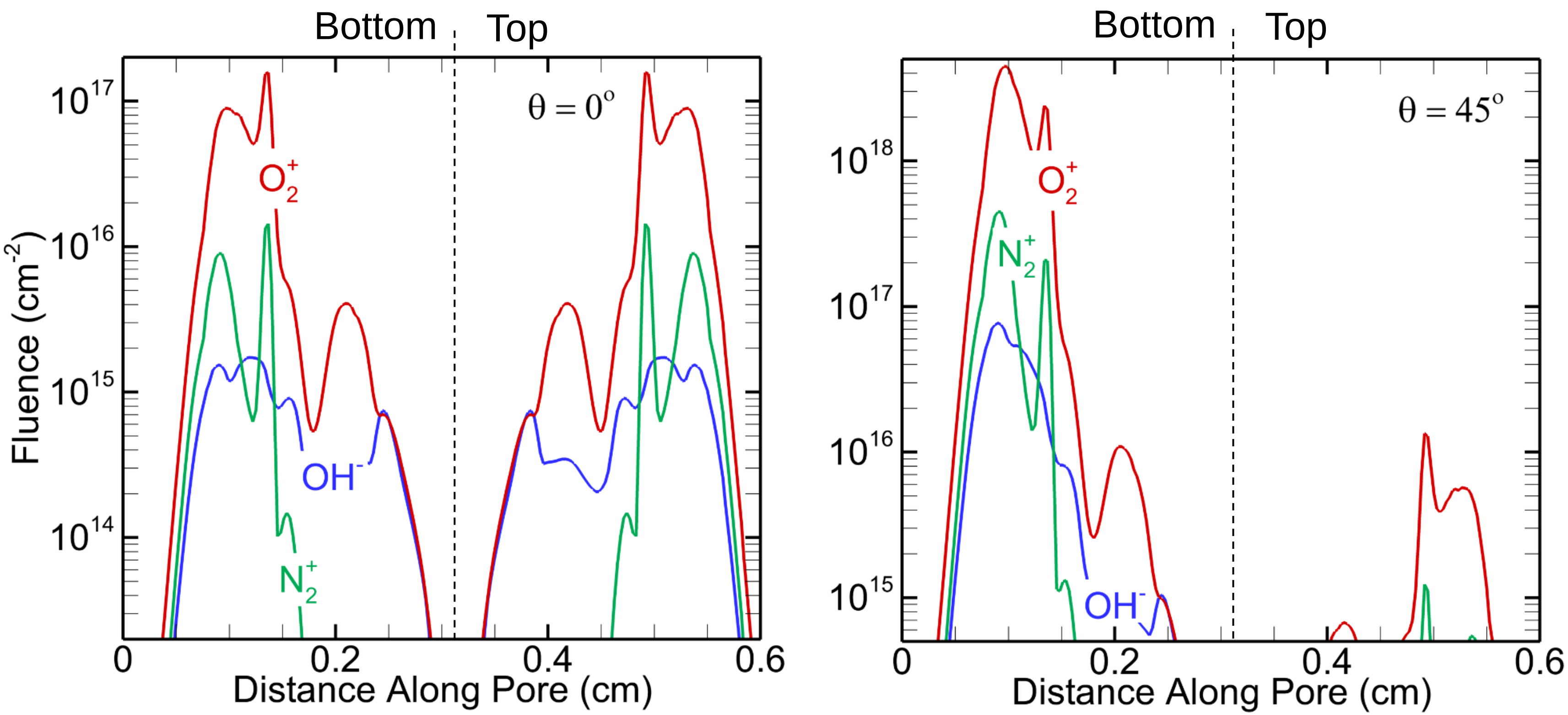
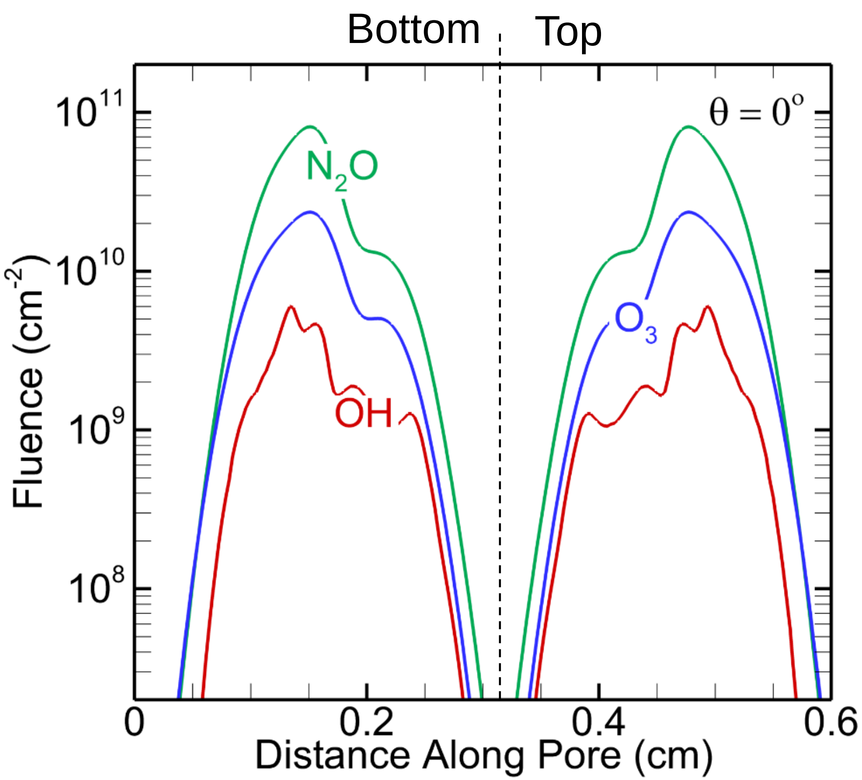
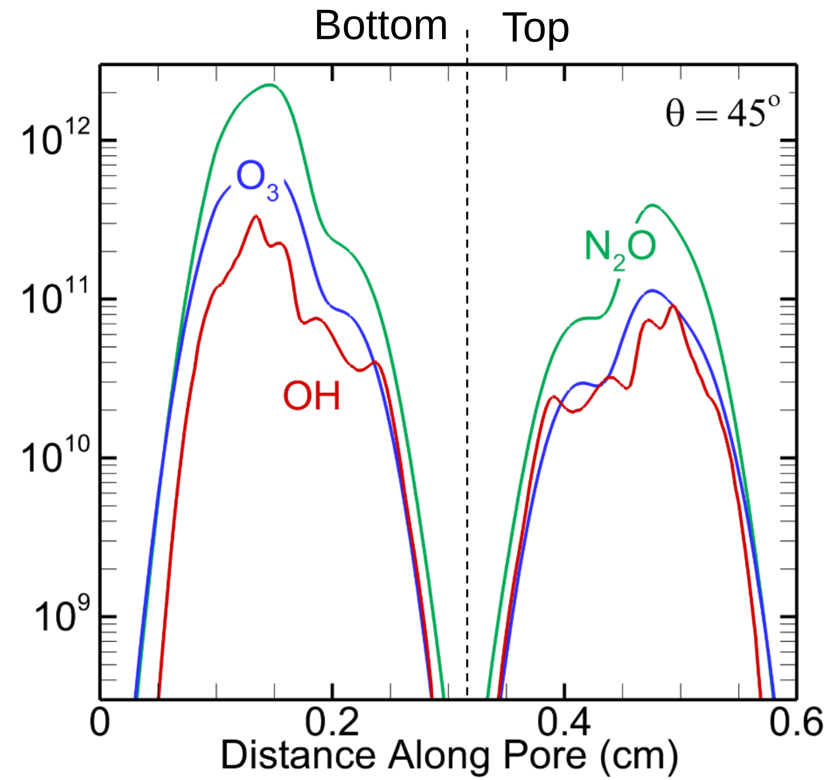
FLUENCES: IONS



- **Distribution of fluences of selected ions along pore walls for incline angles of 0 and 45 degrees at t = 10 ms.**
- **Symmetric fluences to both walls at θ = 0 deg.**
- **Ions recombine within microseconds, fluences drop.**
- **Pulse-on E-fields dictate final distributions of fluences.**

FLUENCES: RONS





- **Distribution of fluences of long-term species along pore walls for incline angles of 0 and 45 degrees at t = 10 ms.**
- **With time, asymmetry relaxes – species react in the gas-phase and fluxes of neutral RONS are driven by diffusion.**

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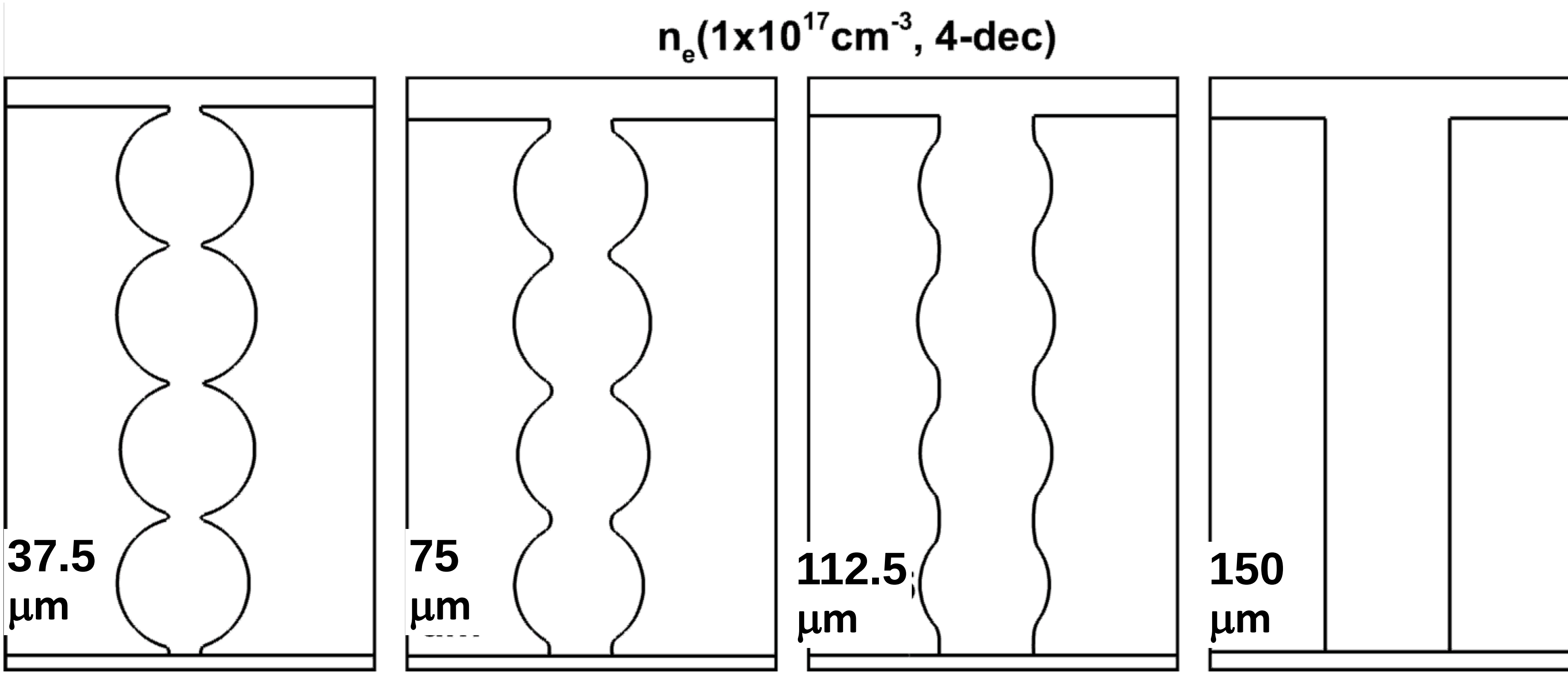
PORE OPENING – WAVY IN THE VERTICAL



- Larger pore opening results in less shadowing and less prominent restrike SIW.
- With opening larger than SIW thickness, SIWs become independent. Center of "pore" is not filled with plasma.
- 1 atm, humid air, -8 kV , 5 ns

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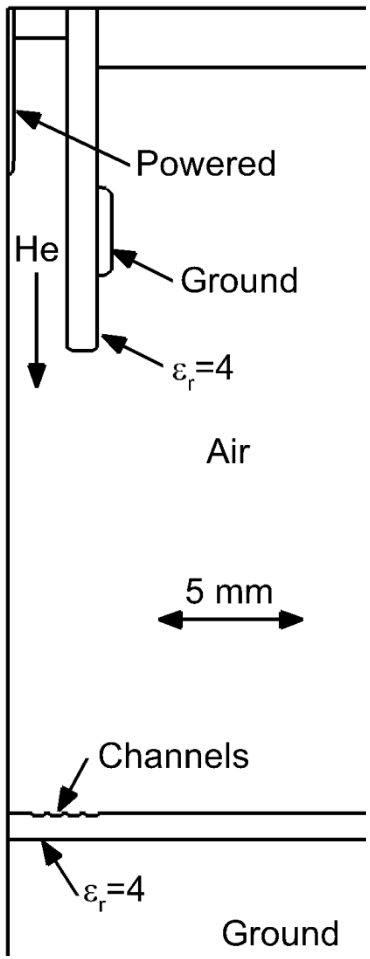
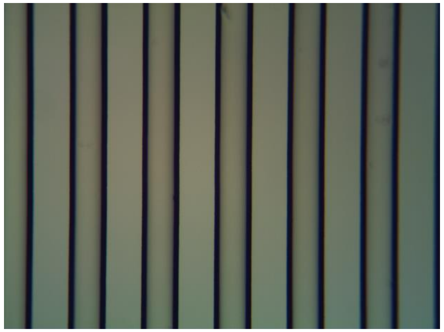
JETS ON STRIPES

JET INTERACTIONS WITH CHANNELS





- **UMD fabricated oxide-covered channels in Si**

- **Prelude to experiments performed with standard surfaces**
- **He/5 ppm ns pulsed plasma jet into humid air incident onto channels (-25 kV, 25 ns)**
- **Channels have varying width, spacing depth.**

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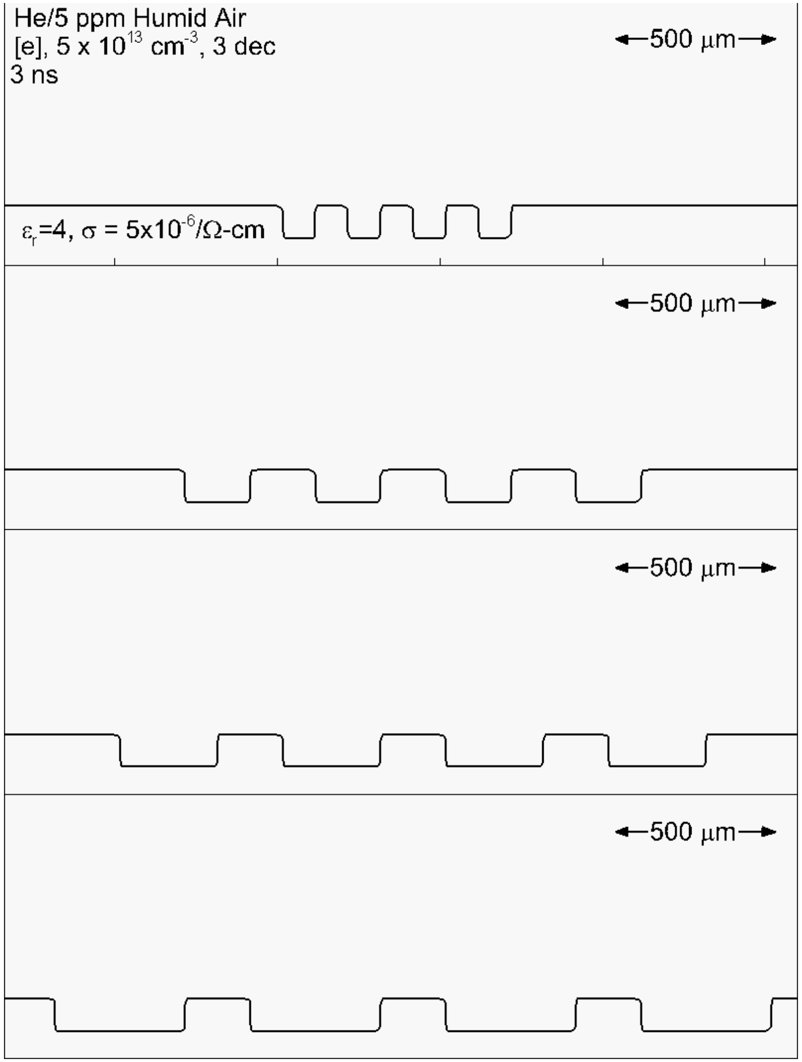
CHANNEL SPACING: PLASMA FILLING [e]



- He/5 ppm air imp. ns pulsed plasma jet into humid air incident onto channels (-25 kV, 25 ns)
- [e], 5 x 10^{13} cm^{-3} (3 dec)
- Channels depth constant (100 μm) with varying width.
- Ionization wave (IW) is attracted to apex of channels due to larger electric field above high permittivity material (smaller capacitance)
- Charging of apex diverts plasma into channels.
- Channels exceeding 250-300 μm do not uniformly fill.

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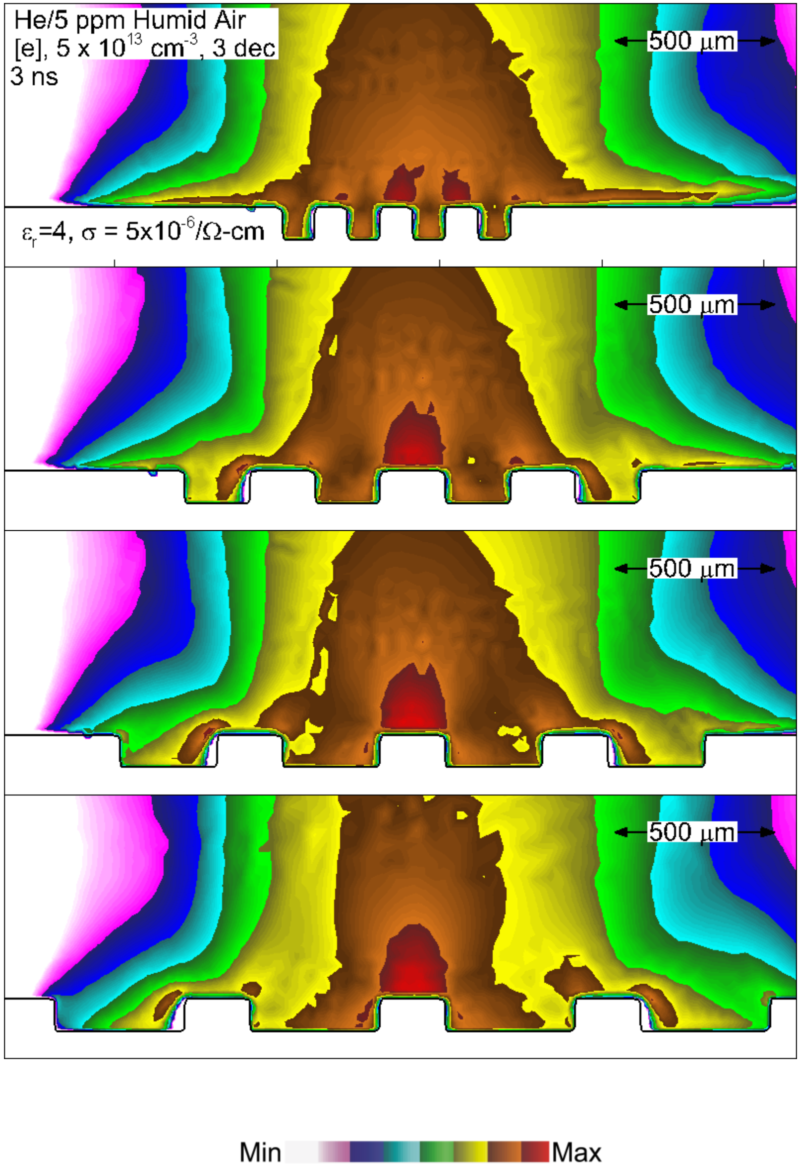
CHANNEL SPACING: PLASMA FILLING [e]



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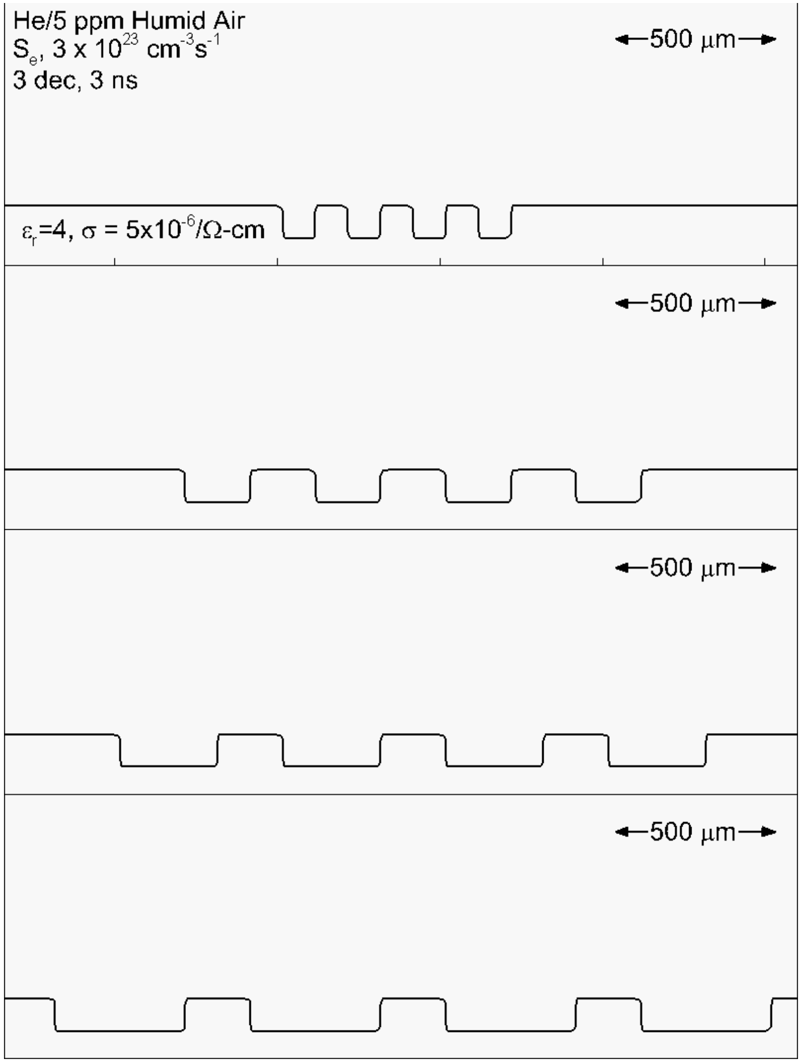
CHANNEL SPACING: TRANSITION TO SIW - S_e



- **He/5 ppm ns pulsed plasma jet into humid air incident onto channels (-25 kV, 25 ns)**
- **Ionization source 3 x 10^{23} cm^{-3} s^{-1} (3 dec)**
- **IW penetrates the depth and fills narrower channels.**
- **Wider channels support small surface ionization waves.**
- **Electric field enhancement at corners maintains small sources after SIW passes – discharging of capacitance.**

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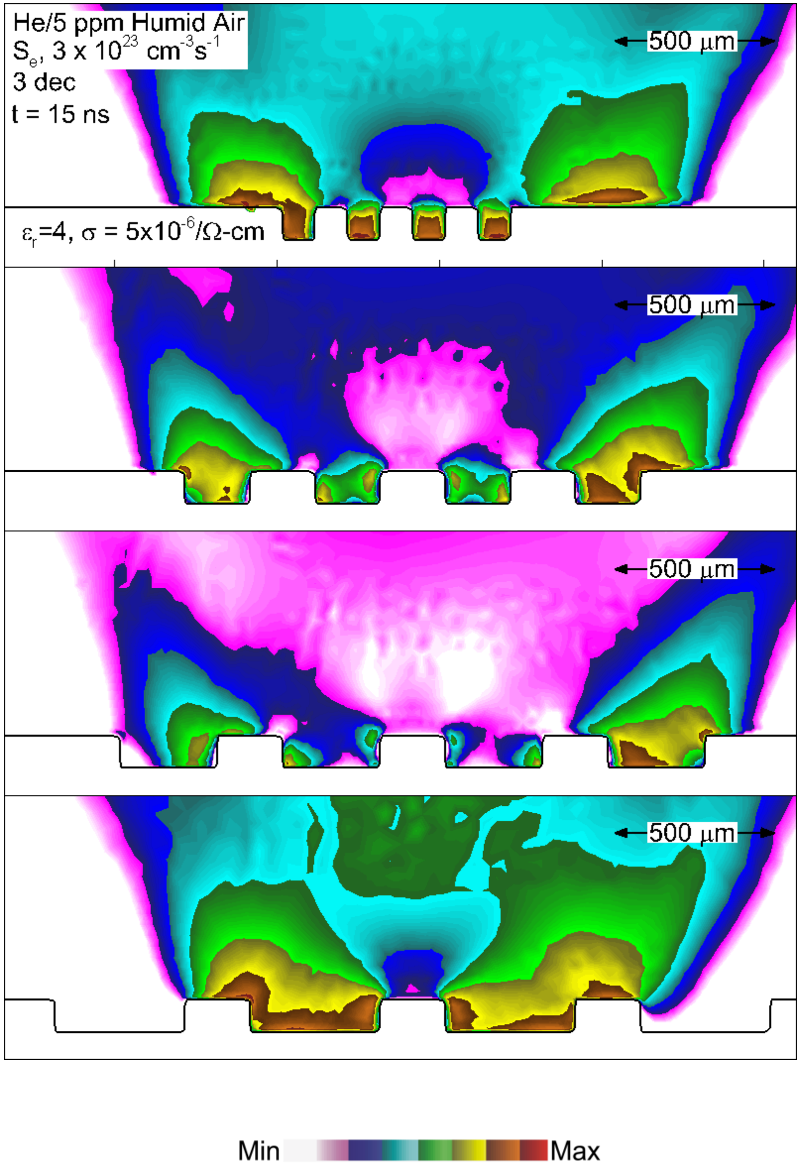
CHANNEL SPACING: TRANSITION TO SIW - S_e



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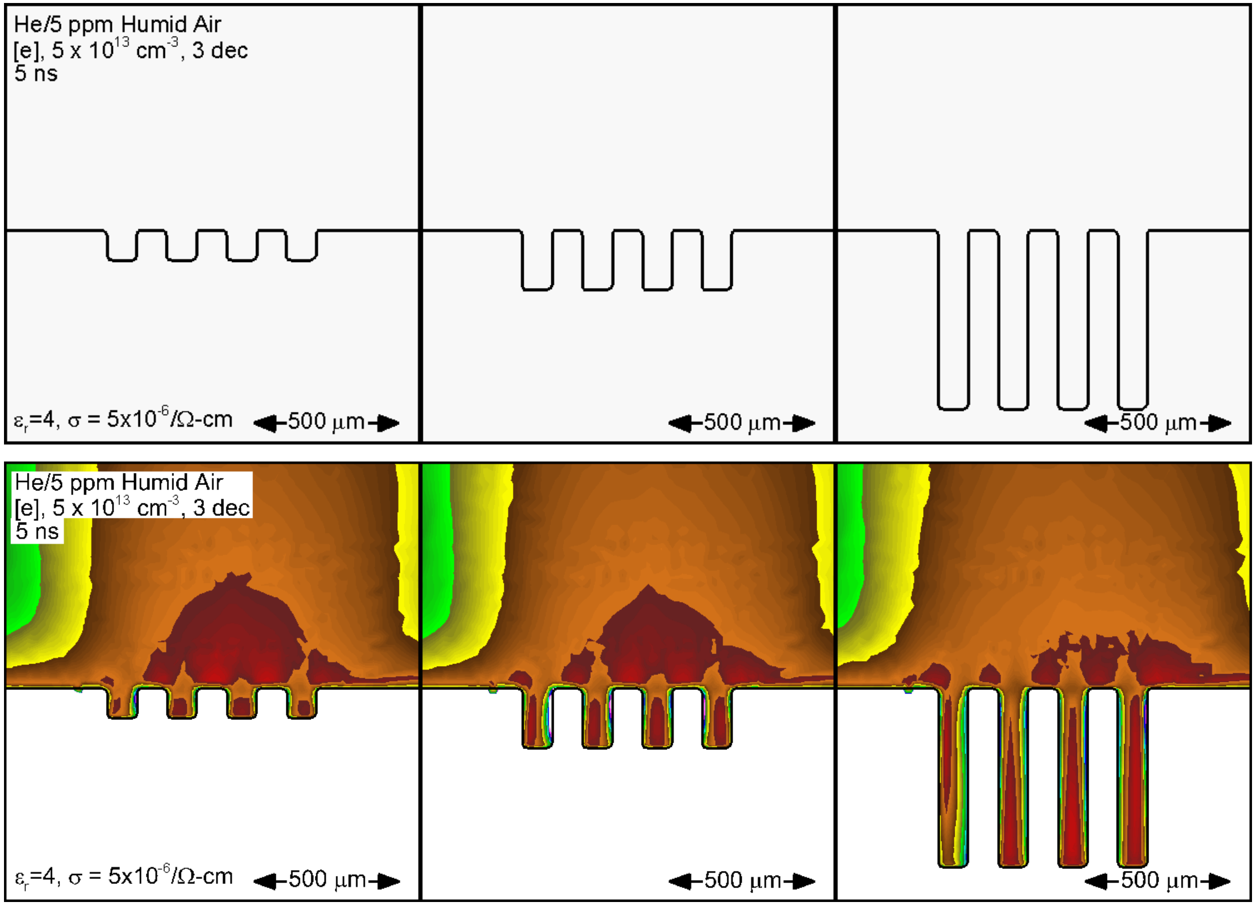
CHANNEL DEPTH-ELECTRON DENSITY



- He/5 ppm plasma jet into incident onto channels (-25 kV, 25 ns)
- Electron density 5 x 10^{13} cm^{-3} (3 dec)
- IW penetrates channels – as SIW for deeper channels.
- Charging of channels provides energy source to sustain ionization.
- Channels see each other, pushing plasma to opposite walls.

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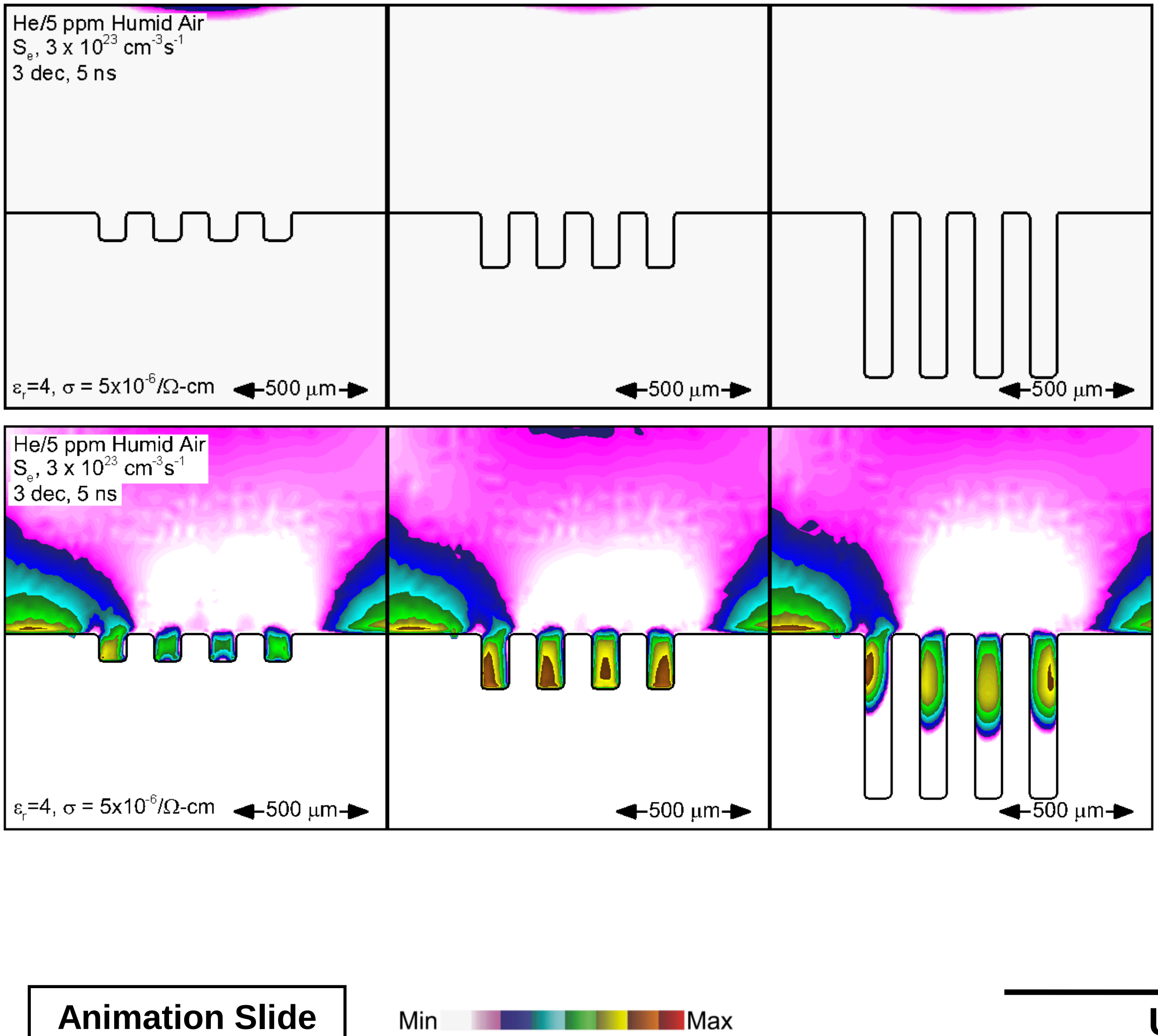
CHANNEL DEPTH-IONIZATION SOURCE



- **He/5 ppm plasma jet into incident onto channels (-25 kV, 25 ns)**
- **Ionization source 3 x 10^{23} $cm^{-3}s^{-1}$ (3 dec)**
- **IW penetrates channels.**
- **Aspect ratio > 1, transition to SIW.**
- **Charging of channels provides energy source to sustain ionization.**
- **Channels see each other, pushing SIW to opposite walls.**

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CONCLUDING REMARKS

- **Geometrically and materially complex surfaces have synergistic interactions with atmospheric pressure plasmas (APPs)**
- **The short mean free paths of electrons and ions localize surface complexities, that in turn intensify feedback.**
- **The surface-hugging nature of SIWs can be compromised by surface roughness that (combined with high ε_r) produces critically large local field enhancement that initiates hopping.**
- **Surface charging is not independent of topology and is responsible for spatially dependent restrike SIWs…**
- **…And the relationship between APPs and complex dielectric surfaces is all about charging.**
- **Reactant fluxes of neutral reactive species are far more uniform than ions.**
- **However, the topology dependent differential charging of surfaces likely plays a role in catalytic reactivity of those neutrals.**