Univ. Madeira

Modelling low-current quasi-stationary gas discharges: mathematical aspects and a practical guide

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Online Low Temperature Plasma (OLTP) seminar

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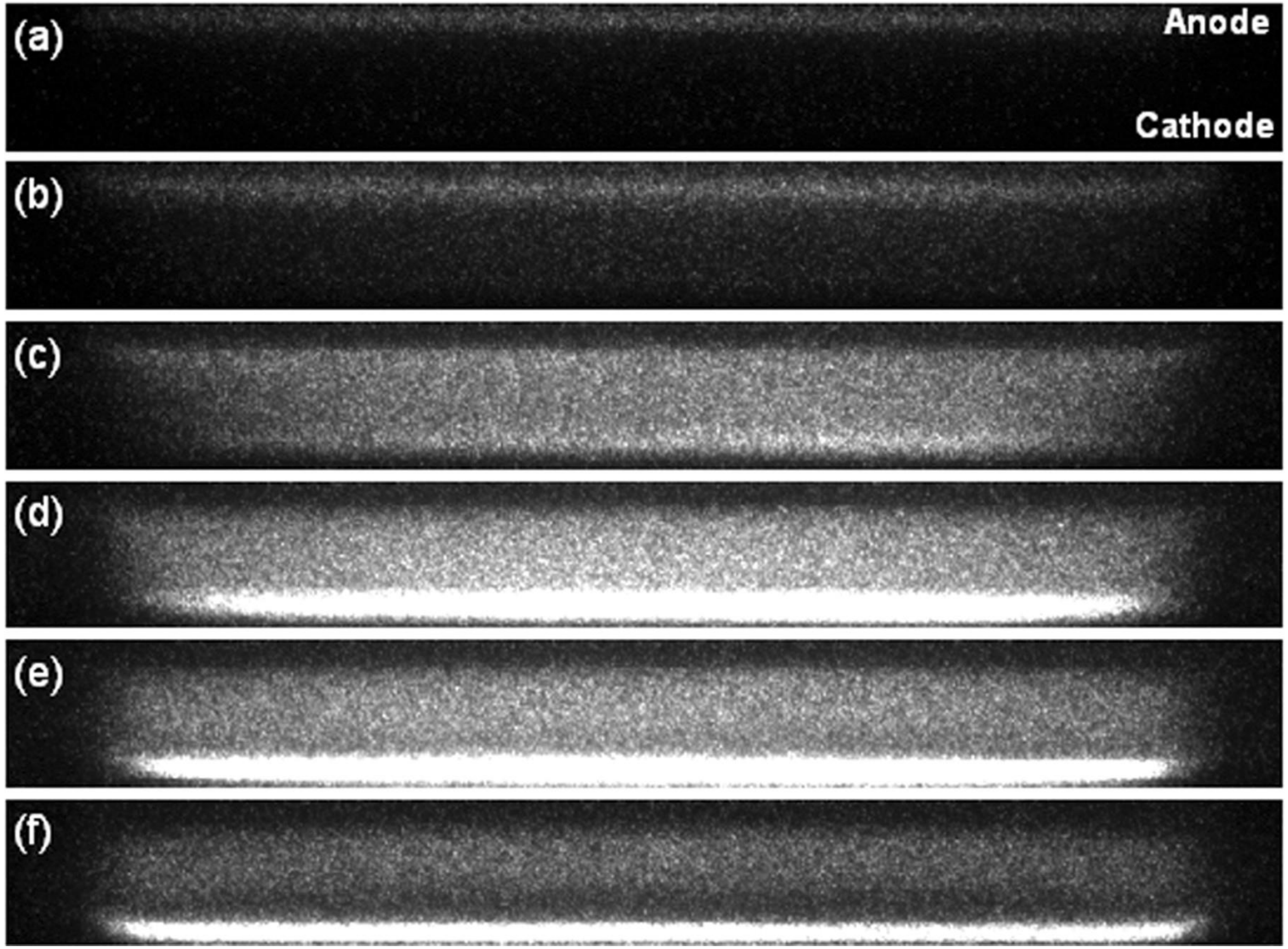
Weakly nonuniform electric field: Townsend discharges

Townsend discharge

- The electric field is uniform.
- The avalanche mechanism: n_e exponentially increases from the cathode to the anode.
- The light intensity is maximal near the anode.

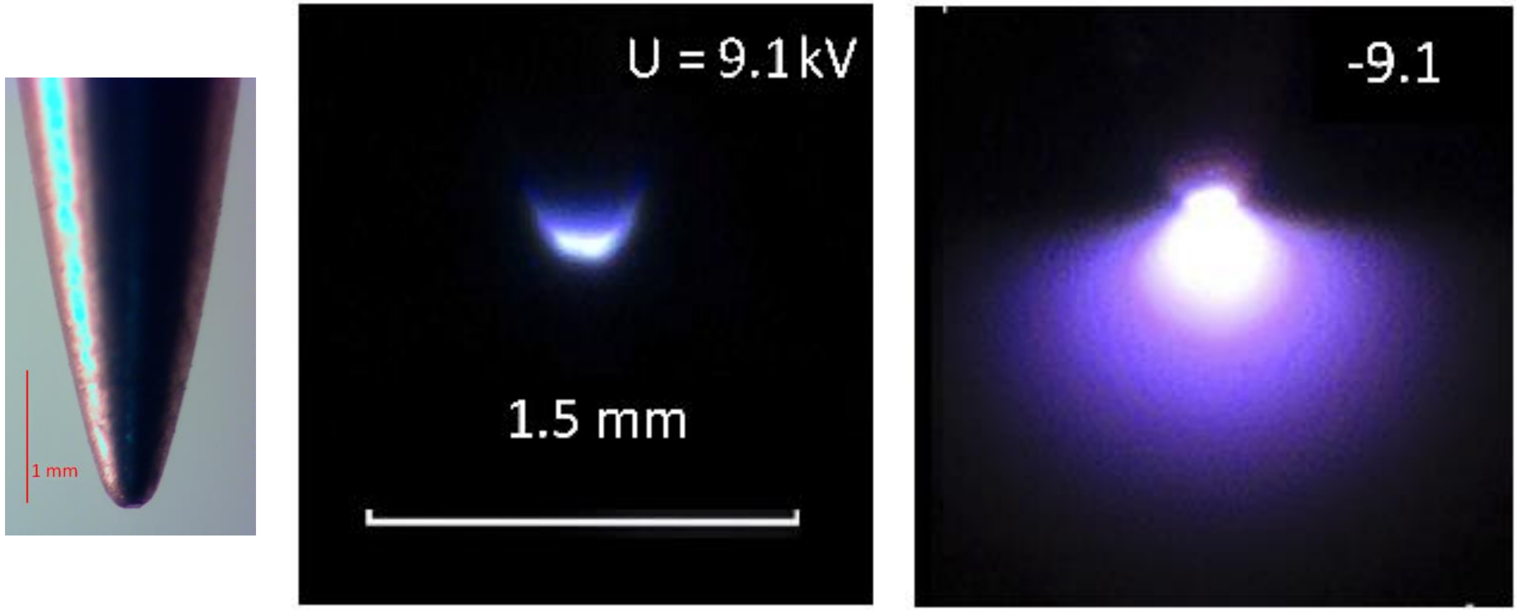
Glow discharge

- The electric field is highly non-uniform.
- Anode dark space, positive column, Faraday dark space, and negative glow.

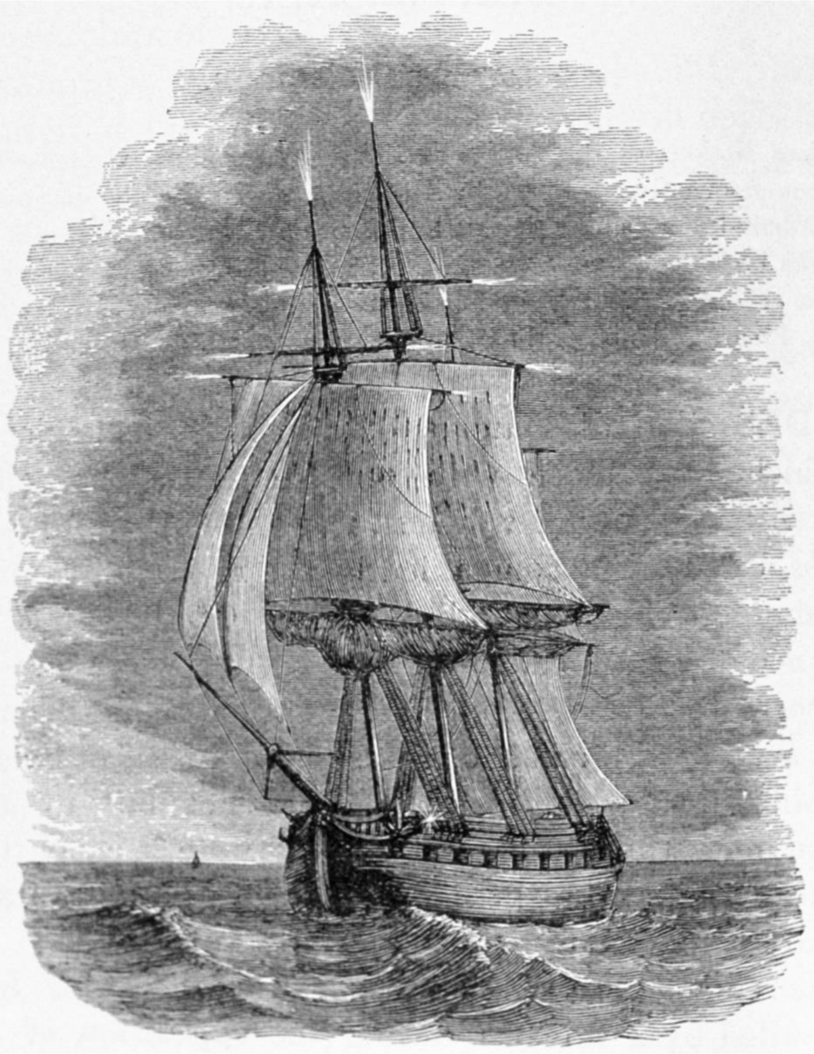


Transition from a Townsend discharge to a glow discharge. He, 1 atm. (a): t=0. (b): 200 ns. (c): 320 ns. (e): 460 ns. (f): 540 ns. From Luo *et al*, Appl. Phys. Lett. **91**, 221504 (2007).

Strongly nonuniform electric field: corona discharges



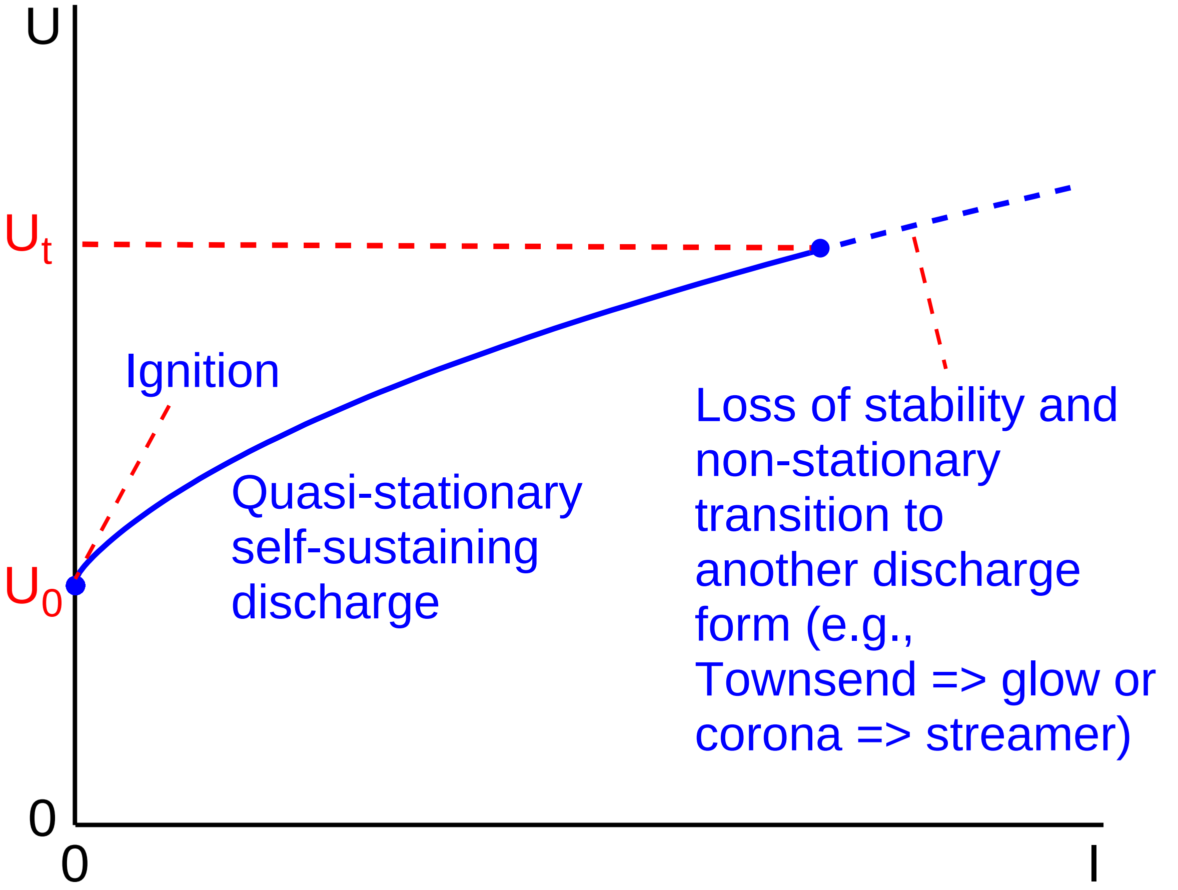
Corona discharge in pin-to-plane geometry. Air, 1 atm. From Ferreira *et al,* IEEE Trans. Plasma Sci. **48**, 4080 (2020).



St. Elmo's Fire on Masts of Ship at Sea, in The Aerial World, by G. Hartwig, London, 1886. Source: https://en.wikipedia.org/wiki/St._Elmo%27s_fire#/media/File:Elmo's_fire-2.jpg

The aim: to compute a quasi-stationary discharge in the whole range of existence

- Three steps:
  - Modelling of **the ignition of a self-sustaining discharge**;
  - Modelling of the **quasi-stationary evolution of the discharge with increasing current**;
  - The determination of the **current range where the quasi-stationary discharge becomes unstable** and is disrupted.



- Each of these three steps is considered in some detail with examples, referring mostly to discharges in atmospheric-pressure air.

Outline of the talk

- Introduction: low-current quasi-stationary gas discharges
- Model of low-current discharges in high-pressure air
- Computation of the initiation of self-sustaining gas discharges
- Modelling of quasi-stationary evolution of the discharge with increasing current
  - Stationary vs. time-dependent solvers
  - Integrated modelling of quasi-stationary low-current discharges
- The determination of the current range where the quasi-stationary discharge becomes unstable
- Conclusions

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Model of low-current discharges in high-pressure air

Equations

Conservation and transport equations for charged particles, excited states, and radicals (drift-diffusion local-field approximation)

$$\frac{\partial n_x}{\partial t} + \nabla \cdot \mathbf{J}_x = S_x$$

$$\mathbf{J}_x = -D_x \nabla n_x - Z_x \mu_x n_x \nabla \varphi$$

Poisson equation

$$\varepsilon \nabla^2 \varphi = -e \sum_x Z_x n_x$$

Equations for the photoionization rate

$$S_{ph} = \sum_{i=1}^{3} S_{ph}^{(i)} \qquad \nabla^2 S_{ph}^{(i)} - \left(\lambda_i p_{O_2}\right)^2 S_{ph}^{(i)} = -A_i p_{O_2}^2 \xi I,$$

A_i, λ_i: parameters of the three-exponential fit function.

ξ: probability of ionization of a molecule on photon absorption.

I: local photon production rate.

Boundary conditions

Anode

$$\frac{\partial n_{n,e}}{\partial n} = 0$$

$$\left(J_p\right)_n = -\frac{n_p}{2}\sqrt{\frac{8k_B T_h}{\pi m_p}}$$

$$\varphi = U$$

$$S_{ph}^{(i)} = 0$$

Cathode

$$\left(J_n\right)_n = -\frac{n_n}{2}\sqrt{\frac{8k_B T_h}{\pi m_n}}$$

$$\left(J_e\right)_n = J_{em}^{(e)} - \frac{n_e}{2}\sqrt{\frac{8k_B T_e}{\pi m_e}}$$

$$\frac{\partial n_p}{\partial n} = 0$$

$$\varphi = 0$$

$$S_{ph}^{(i)} = 0$$

Numerical boundaries

$$\frac{\partial n_x}{\partial n} = 0 \qquad \frac{\partial \varphi}{\partial n} = 0 \qquad \frac{\partial S_{ph}^{(i)}}{\partial n} = 0$$

$J_{em}^{(e)}$: electron emission flux (all relevant mechanisms).

m_x: particle mass of species x.

Model of low-current discharges in high-pressure air

"Minimal" kinetic scheme for low-current discharges in high-pressure air

Reaction	References for kinetic coefficients
$e + M \Rightarrow 2e + A^+$	Ferreira *et al*, J. Phys. D: Appl. Phys. **52**, 355206 (2019)
$e + O_2 \Rightarrow O^- + O$	Ferreira *et al*, J. Phys. D: Appl. Phys. **52**, 355206 (2019)
$e + O_2 + M \Rightarrow O^-_2 + M$	Sigmond, J. Appl. Phys. **56**, 1355 (1984)
$O^-_2 + O_2 \Rightarrow e + 2O_2$	Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021)
$O^- + N_2 \Rightarrow e + N_2O$	Pancheshnyi, J. Phys. D: Appl. Phys. **46**, 155201 (2013)
$O^- + O_2 \Rightarrow O + O^-_2$	Pancheshnyi, J. Phys. D: Appl. Phys. **46**, 155201 (2013)
$O^- + O_2 + M \Rightarrow O^-_3 + M$	Pancheshnyi, J. Phys. D: Appl. Phys. **46**, 155201 (2013)
$M + h\upsilon \Rightarrow e + A^+$	Bourdon *et al*, Plasma Sources Sci. Technol. **16**, 656 (2007)
$A^+ + B^- \Rightarrow$ products	Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021)
$A^+ + e \Rightarrow$ products	Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021)

A^+: the effective positive ion species. B^-: any of the negative ions O^-, O^-_2, and O^-_3. M: any of the molecules N_2 and O_2. From Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021).

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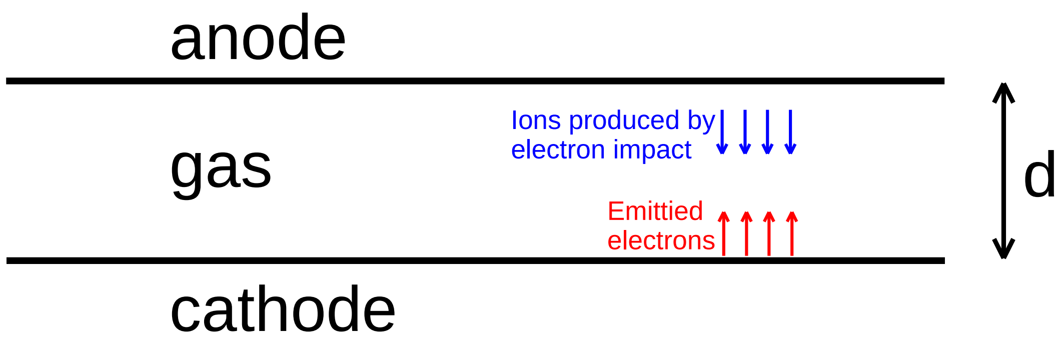
Computing initiation of self-sustaining gas discharges

- The initiation of a self-sustaining discharge: **the applied voltage is just sufficient for the electron impact ionization and the photoionization to compensate the electron losses.**
- Parallel-plate electrodes, one ion species, diffusion of the charged particles and photoionization are neglected: **Townsend criterion**



$$\alpha d = \ln(1/\gamma+1)$$

α: ionization coefficient

γ: secondary electron emission coefficient

- **No simple solution for other cases** (e.g., with photoionization or in the presence of dielectric surfaces).
- **The problem of ignition of self-sustaining discharges may be solved by accurate mathematical means and this solution is relatively simple.**

Computing initiation of self-sustaining gas discharges

At the ignition, the charged particle densities are low =>

- **The electric field is not perturbed** by space charges,

$$\nabla^2 \varphi = 0,$$

and may be determined by means of standard electrostatic simulations.

- **No need to consider nonlinear and multistep processes**, e.g., reactive collisions involving two or more particles produced in the discharge (ions, electrons, excited states, or radicals) =>

- No stepwise ionization, no recombination, no need to consider excited states and radicals =>

- **It is sufficient to consider the equations governing the charged particle distributions in the gap.**

Computing initiation of self-sustaining gas discharges

- The ignition voltage is the value of the applied voltage U such that the applied electric field produces an ionization intensity just sufficient to compensate for the losses of charged particles, so **a steady-state balance of the charged particles is reached** => The conservation equations should be considered in the stationary form:

$$\nabla \cdot \boldsymbol{J}_x = S_x, \qquad x = e, p, n$$

$$\mathbf{J}_x = -D_x \nabla n_x - Z_x \mu_x n_x \nabla \varphi$$

- This is a **linear homogeneous boundary-value problem, which admits a trivial solution** $n_x = 0$ for all values of U (no discharge has been ignited).

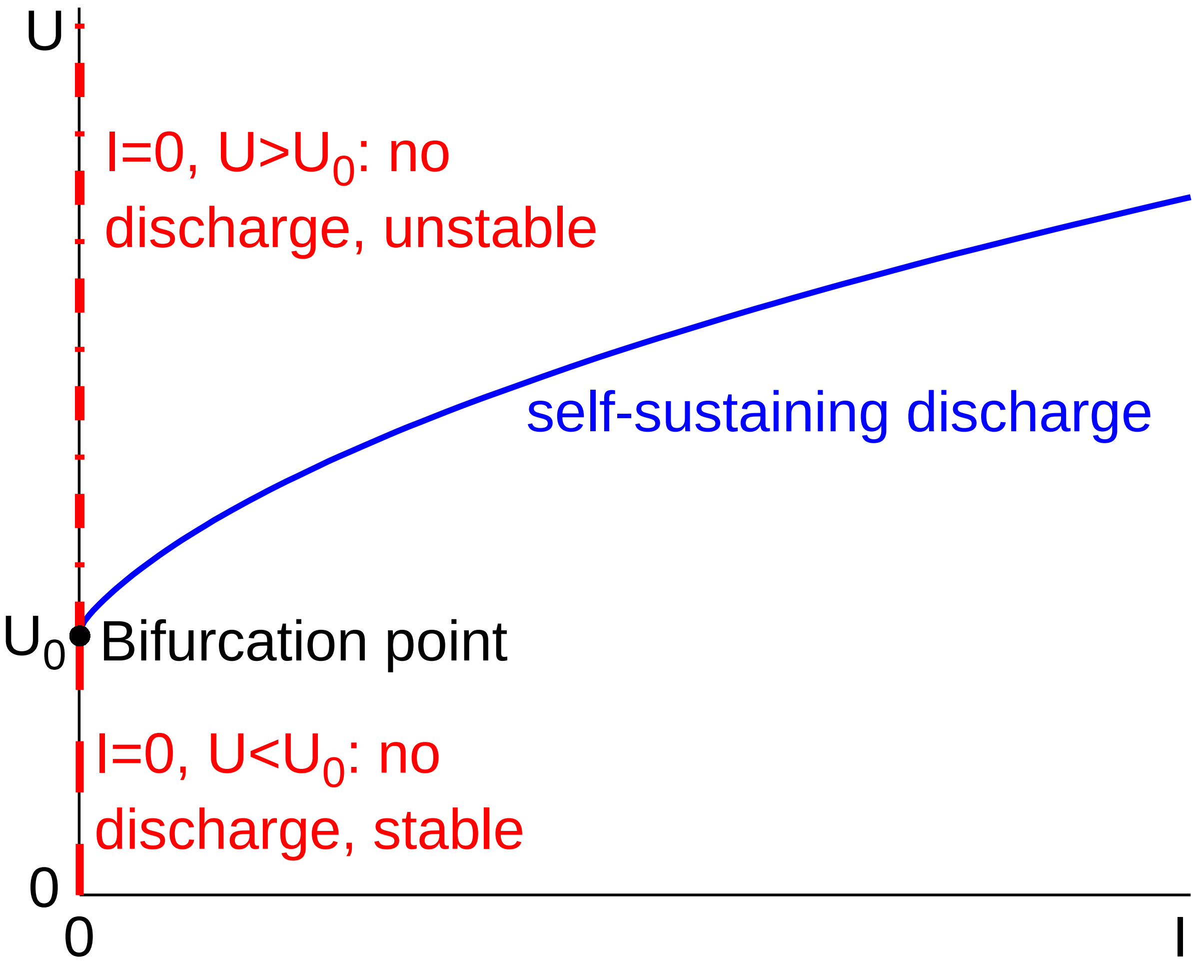
- The aim is to find **a value of** U **such that the problem admits**, in addition to the trivial solution, also **a nontrivial one**.

- This is **an eigenvalue problem** for a system of PDEs and U is the eigenparameter.

Solving the eigenvalue problem for the self-sustainment voltage

- The most **direct approach** is to apply an **eigenvalue solver** directly to the eigenvalue problem as it was formulated, considering *U* as the eigenparameter. Powerful eigen-value solvers are available, e.g. in COMSOL Multiphysics®.
  Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021)

- **Option 2: investigation of stability of no-discharge solution**. An **eigenvalue solver** is needed.
  1D case: Shibata *et al*, Plasma Sources Sci. Technol. **24**, 055014 (2015).



From Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021).

Solving the eigenvalue problem for the self-sustainment voltage

- **Option 3: the resonance method**
  Almeida *et al*, Plasma Sources Sci. Technol. **29**, 125005 (2020)

$$\nabla \cdot \boldsymbol{J}_x = S_x + W, \quad x = e, p$$

W: an external ionization term (does not depend on the particle densities nor on *U*; specified more or less arbitrarily; absolute values are irrelevant).

- ➢ The obtained problem describes a **non-self-sustained discharge** (same gas and same configura-tion) and is solved for different values of *U* by means of **stationary linear solver**.
- ➢ A resonance should appear when the self-sustainment voltage is attained.

- **The three methods are mathematically equivalent** as far as computation of the self-sustainment voltage is concerned =>

Which method is easier to implement and use and which method has wider applicability?

Computing initiation of self-sustaining gas discharges

- **Method 1** (direct solution of the eigenvalue problem for the self-sustainment voltage)

  U=175.05 V, the vertical dashed line.

- **Method 2** (stability of the no-discharge solution)

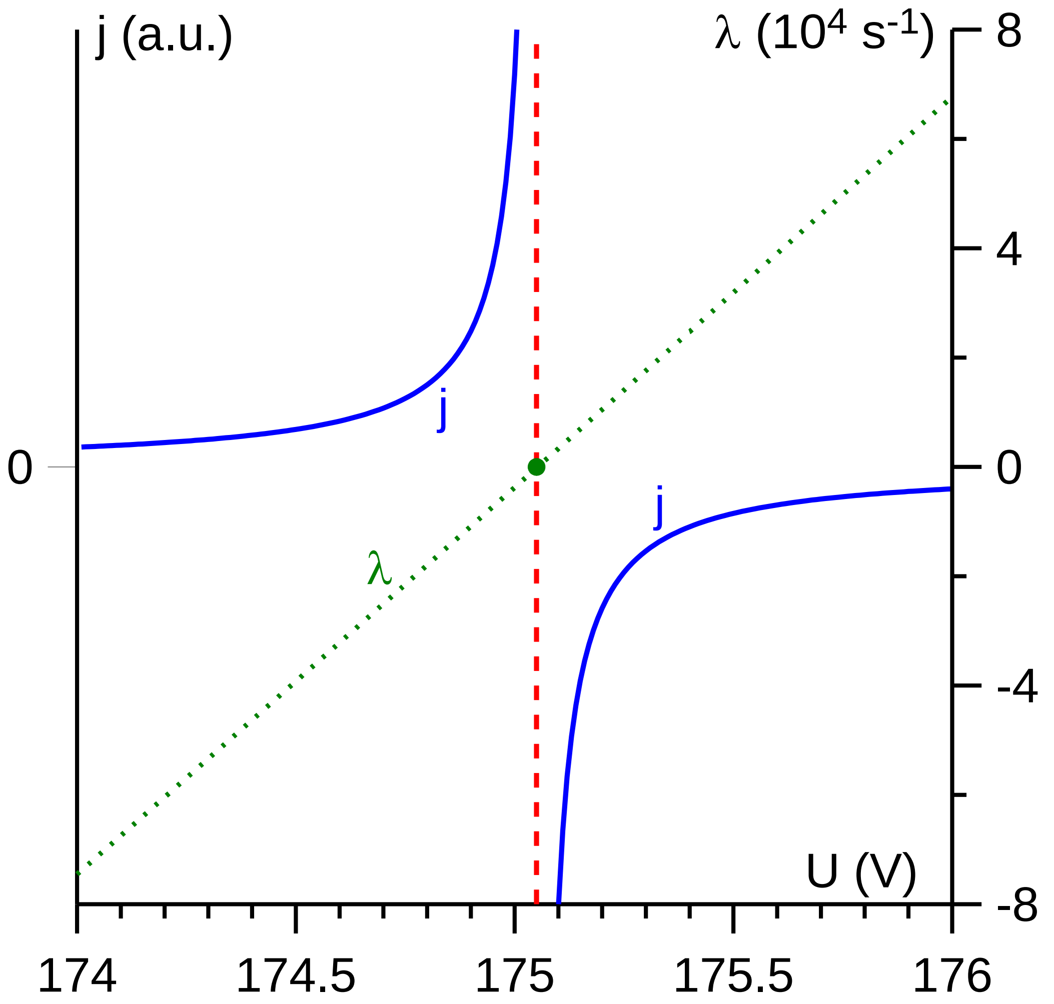
  Dotted: the perturbation growth increment. Circle: the point of neutral stability, *U*=175.05 V.

- **Method 3** (resonance)

  Solid: CDVC of the non-self-sustained discharge. There is a **singularity** at *U*=175.05 V.

  The analytical solution without diffusion is similar, with the singularity at 177.20 V (same as the Townsend-criterion value).

- **The three methods give the same result.**



Parallel-plate discharge in Xe. 30 Torr, gap 0.5 mm, a single ion species, no photoionization, γ = 0.03. From Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021).

Computing initiation of self-sustaining gas discharges

- **Method 1** is direct => requires computing and analysing the spectrum only once.
- **Method 2** requires doing this several times (for different *U* values) => is more laborious.
- **Methods 1 and 2**: finding a spectrum by means of an eigenvalue solver is a nonlinear task and requires care (distinguishing between physical and artificial eigenvalues, mesh dependence, …
- **Method 3** requires solving a stationary boundary-value problem, which is linear => a straightforward solution. The self-sustainment voltage can be readily identified as a singularity in the CVC of the non-self-sustained discharge with the subsequent change of sign of current.

=> Method 3 (the resonance method) is easier to implement and use. It is physics-based, transparent, and provides an initial point for the modelling of quasi-stationary self-sustained discharges.

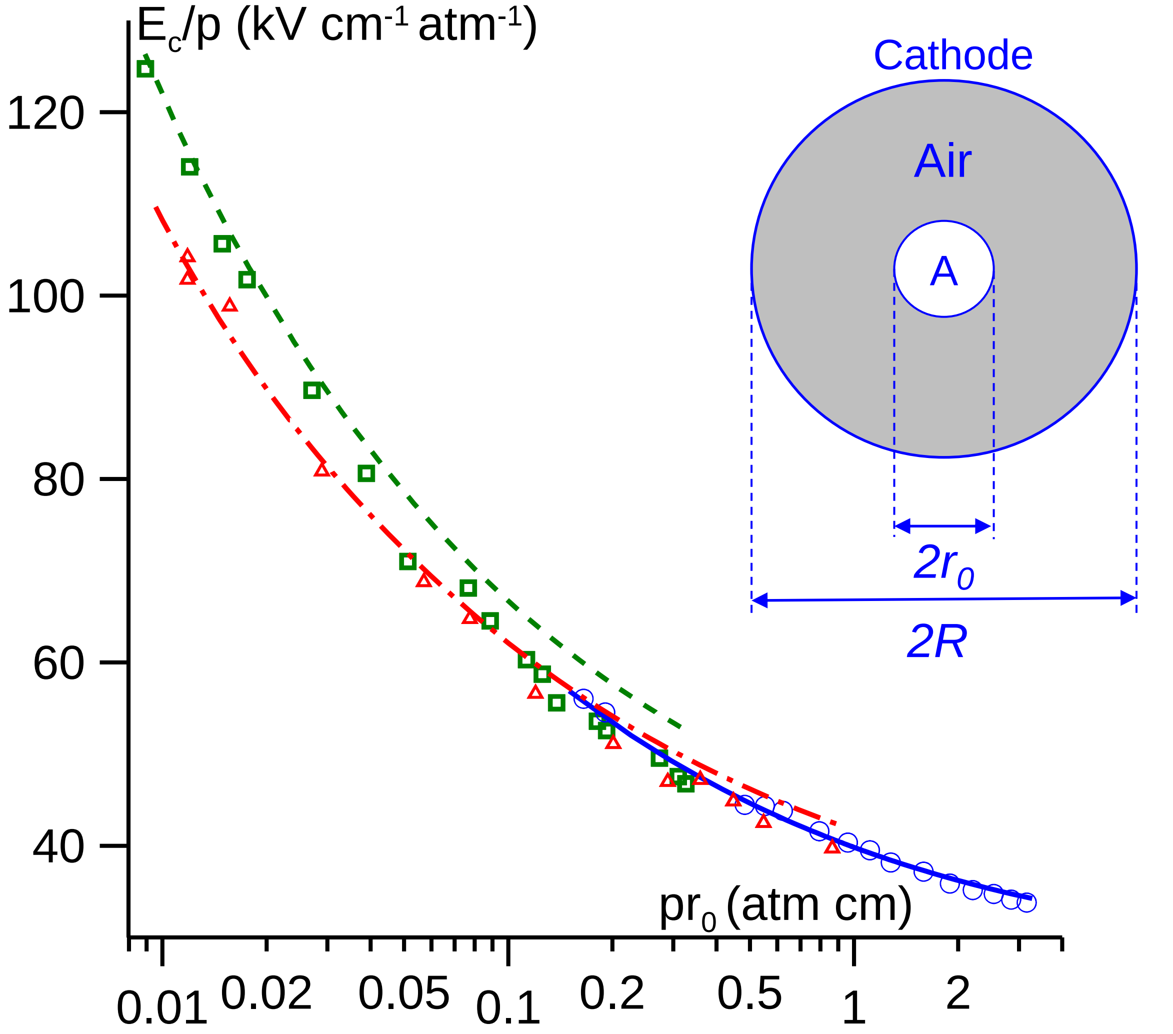
Example: inception of positive corona discharge in air

○, solid: experimental data Waters and Stark, J. Phys. D: Appl. Phys. **8**, 416 (1975) and the modelling.
p = 1 atm, *R* = 29.1 cm, varying r_0.

Δ, dash-dotted: experimental data Robinson, IEEE Trans. Power. Appar. Syst. **86**, 185 (1967) and the modelling.
r_0 = 0.120 cm, *R* = 4.88 cm, varying *p*.

◘, dashed: experimental data Robinson, IEEE Trans. Power. Appar. Syst. **86**, 185 (1967) and the modelling.
r_0 = 0.0089 cm, *R* = 4.88 cm, varying *p*.

The **agreement** between **computed and measured inception field is good**.



Reduced inception field at the surface of positive wire electrode.
From Ferreira *et al*, J. Phys. D: Appl. Phys. **52**, 355206 (2019).

Example: inception of positive corona discharge in air

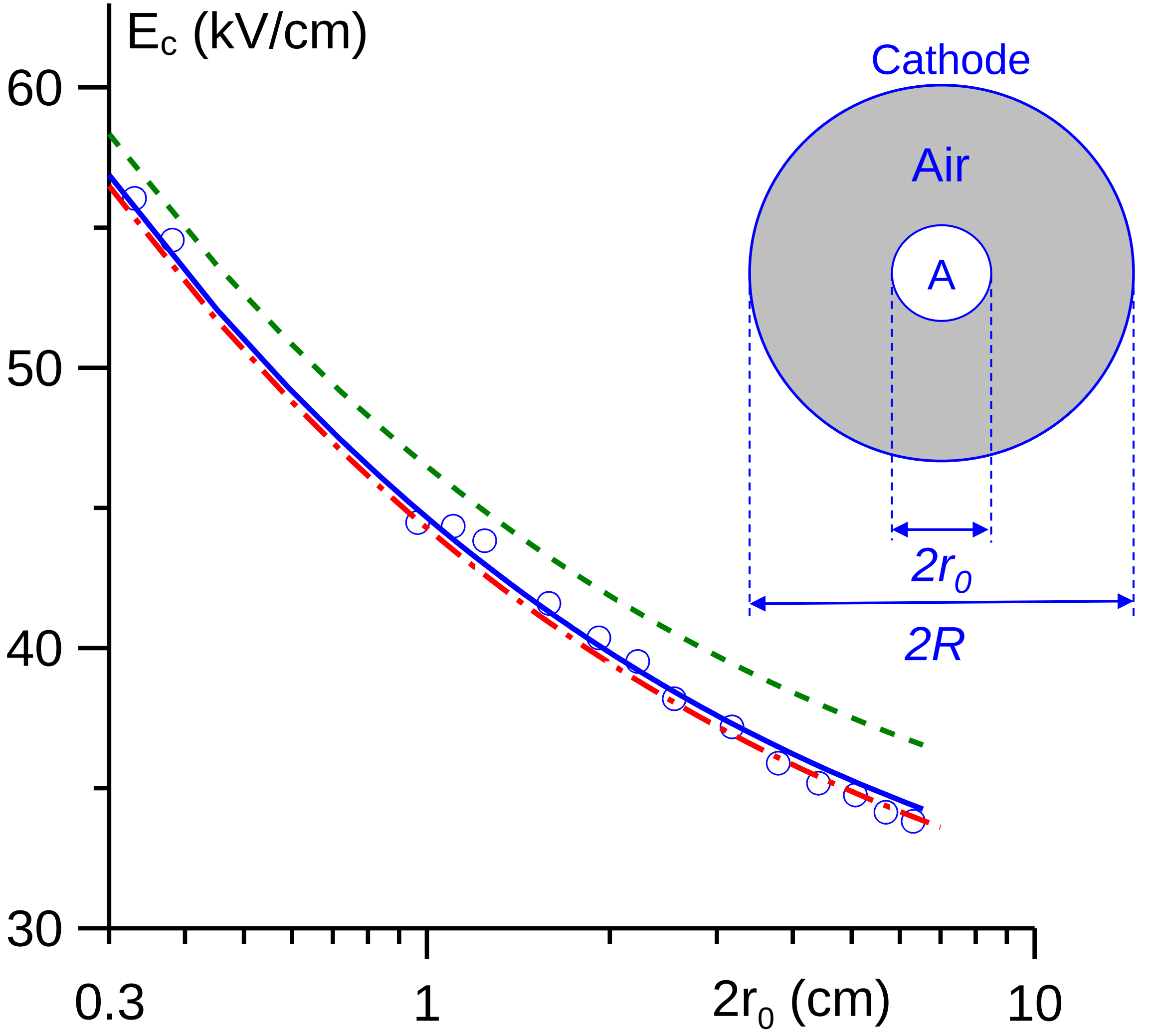
○: experimental data Waters and Stark, J. Phys. D: Appl. Phys. **8**, 416 (1975).
p = 1 atm, R = 29.1 cm, varying r_0.

Solid: full kinetic scheme.

Dashed: kinetic scheme without account of detachment process.

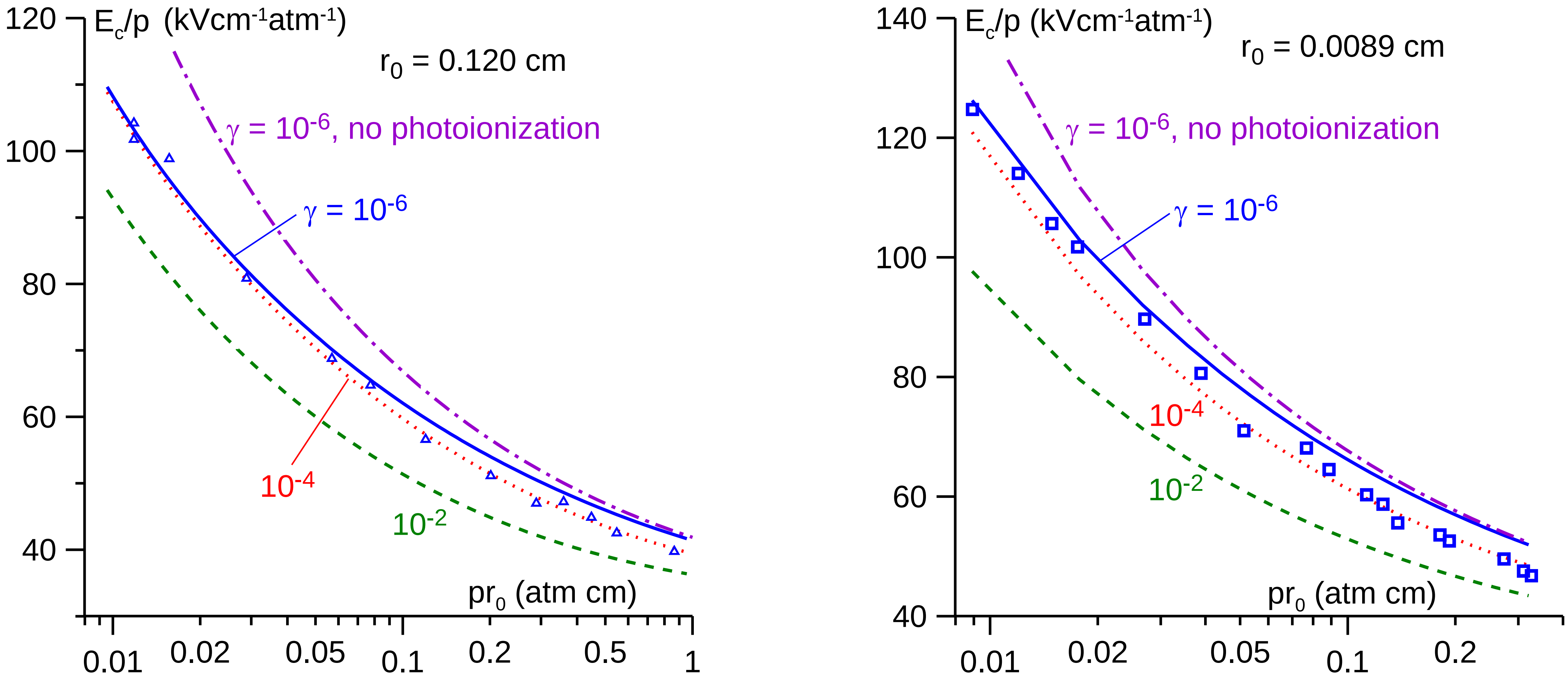
Dash-dotted: kinetic scheme with rates of attachment set to zero.

- Disregard of detachment results in an increase of the inception voltage as it should.
- The dash-dotted line is close to the solid one => **the detachment approximately compensates the attachment.**



Reduced inception field at the surface of positive wire electrode. From Ferreira *et al*, J. Phys. D: Appl. Phys. **52**, 355206 (2019).

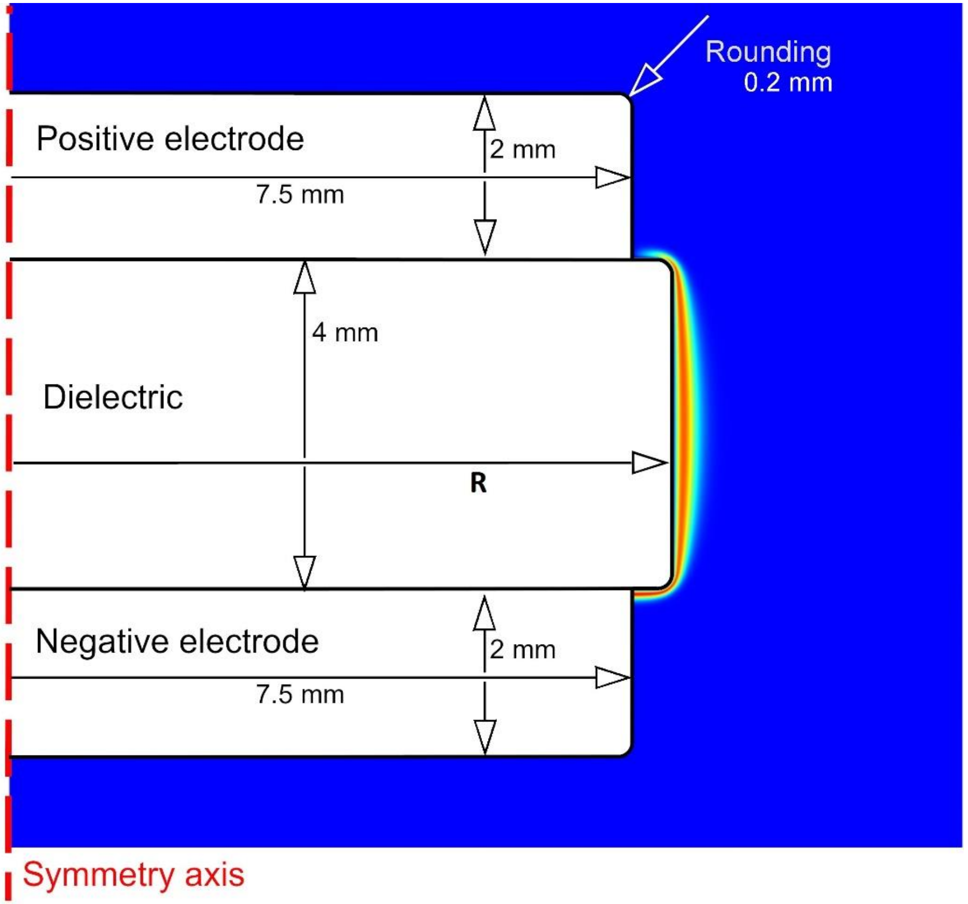
Example: inception of positive corona discharge in air

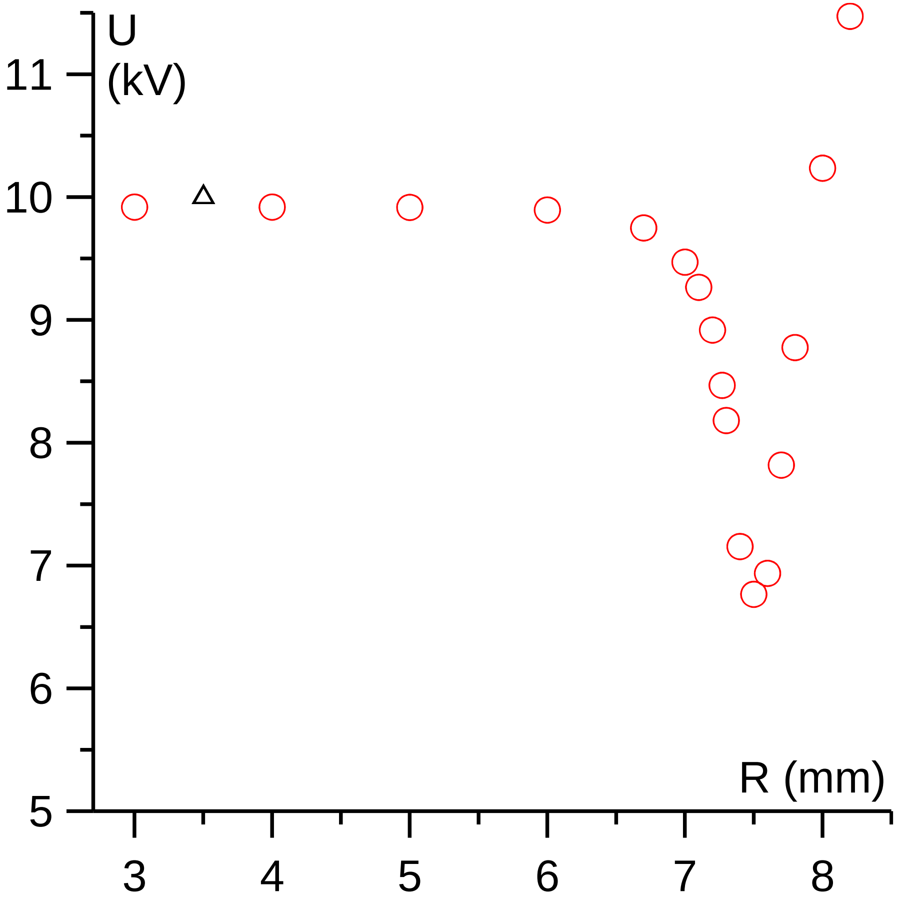


Reduced inception field at the surface of positive wire electrode. From Ferreira *et al*, J. Phys. D: Appl. Phys. **52**, 355206 (2019).

- Points: experimental data Robinson, IEEE Trans. Power. Appar. Syst. **86**, 185 (1967). R = 4.88 cm, varying p.
- Lines: modelling with different values of the secondary electron emission coefficient. The line for $\gamma = 0$ coincides with the line for $\gamma = 10^{-6}$.

Example: inception of discharge along a dielectric surface





Inception voltage of discharge between disc electrodes separated by a cylindrical dielectric. Air, 1 atm. From Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021).

- Circles: modelling.
- Triangle: breakdown voltage, experiment Pillai and Hackam, J. Appl. Phys. **58**, 146 (1985).

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Stationary vs. time-dependent solvers

- **Time-dependent solvers are routinely used in gas discharge modelling**, including in popular ready-to-use toolkits (e.g., nonPDPSIM, Plasma Module of COMSOL Multiphysics).
- Time-dependent solvers can be used for the simulation of **quasi-stationary gas discharges**: an initial state of a discharge is specified and its relaxation over time is followed until a steady state has been attained.
- An alternative is to use **stationary solvers**, which solve steady-state equations by means of an **iterative process unrelated to time relaxation** (usually Newton iterations).
- While being rarely used in gas discharge modelling, stationary solvers do exist, e.g.,:
  - COMSOL Multiphysics® provides stationary solvers for general PDEs;
  - While the Plasma module of COMSOL Multiphysics® is intended to work with time-dependent solvers, it can still be used with stationary solvers;
  - Stationary solvers are provided by Plasimo.

Stationary vs. time-dependent solvers

Stationary solvers offer important advantages in simulations of steady-state discharges:

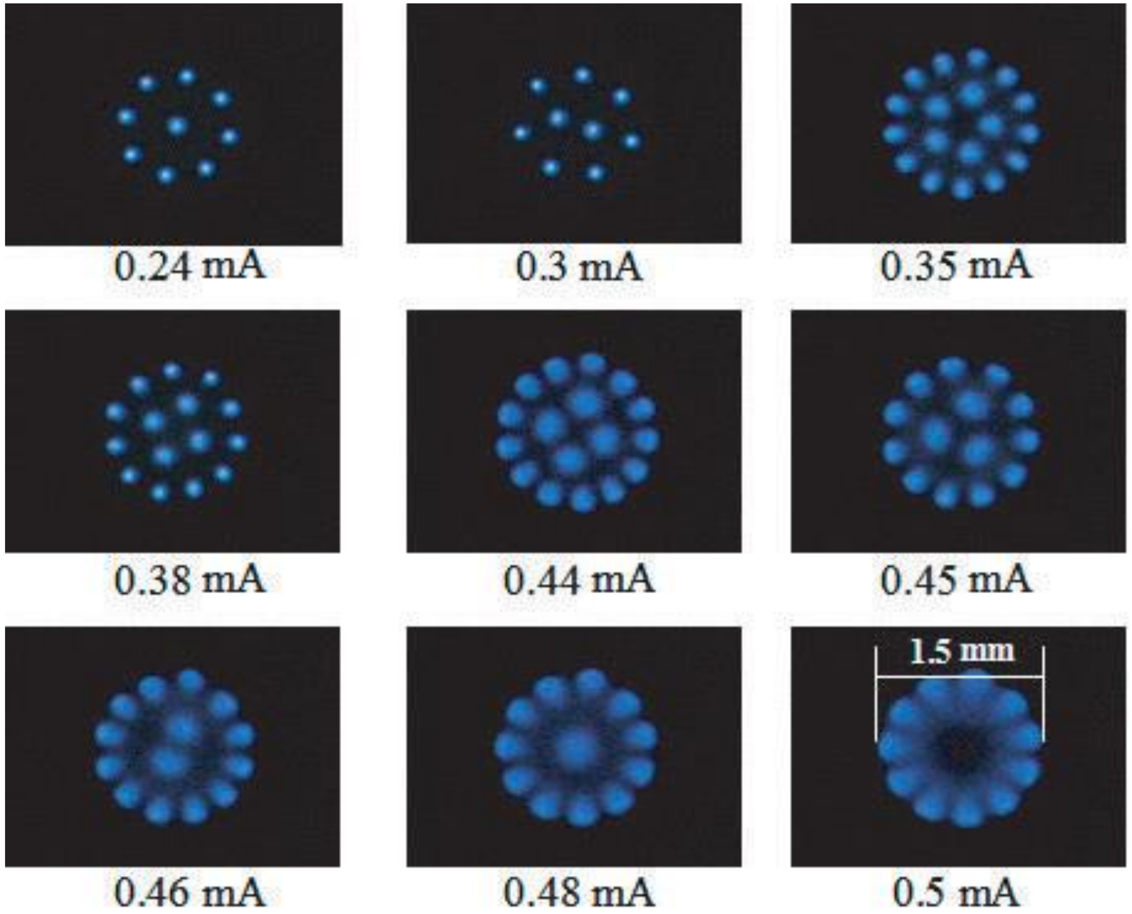
- Stationary solvers are not subject to the Courant–Friedrichs–Lewy criterion or analogous limitations on time stepping => **simulations are much faster, by orders of magnitude in many cases**. (Particularly important for modelling discharges with strongly varying length scales, e.g., corona discharges, where a variation of the mesh element size by orders of magnitude is indispensable.)

- Stationary solvers allow **decoupling of physical and numerical stability**.

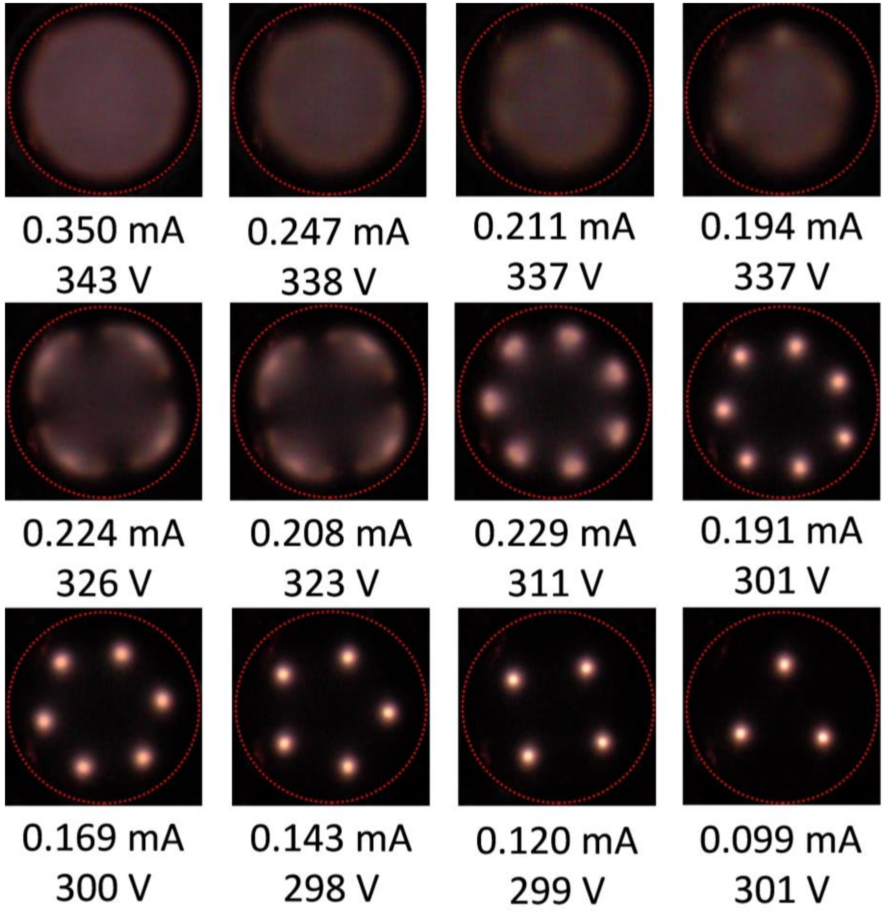
- Stationary solvers are efficient in **computing self-organization**.

- Stationary solvers are efficient in computing **patterns of complex behavior that can manifest itself in the modelling of gas discharges even in apparently simple quasi-stationary situations.**

Self-organization on DC glow cathodes



Glow microdischarge in Xe. 75 Torr, radius 750 μm, height 250 μm. From Schoenbach *et al*, Plasma Sources Sci. Technol. **13**, 177 (2004).



Glow microdischarge in Kr. 150 Torr, radius 375 μm, height 250 μm. From Zhu *et al*, Plasma Sources Sci. Technol. **23**, 054012 (2014).

Self-organization on DC glow cathodes via stationary solvers

Glow microdischarge in Xe. 30 Torr, radius 0.5 mm, height 0.5 mm, diffusion losses neglected.

a): CVCs

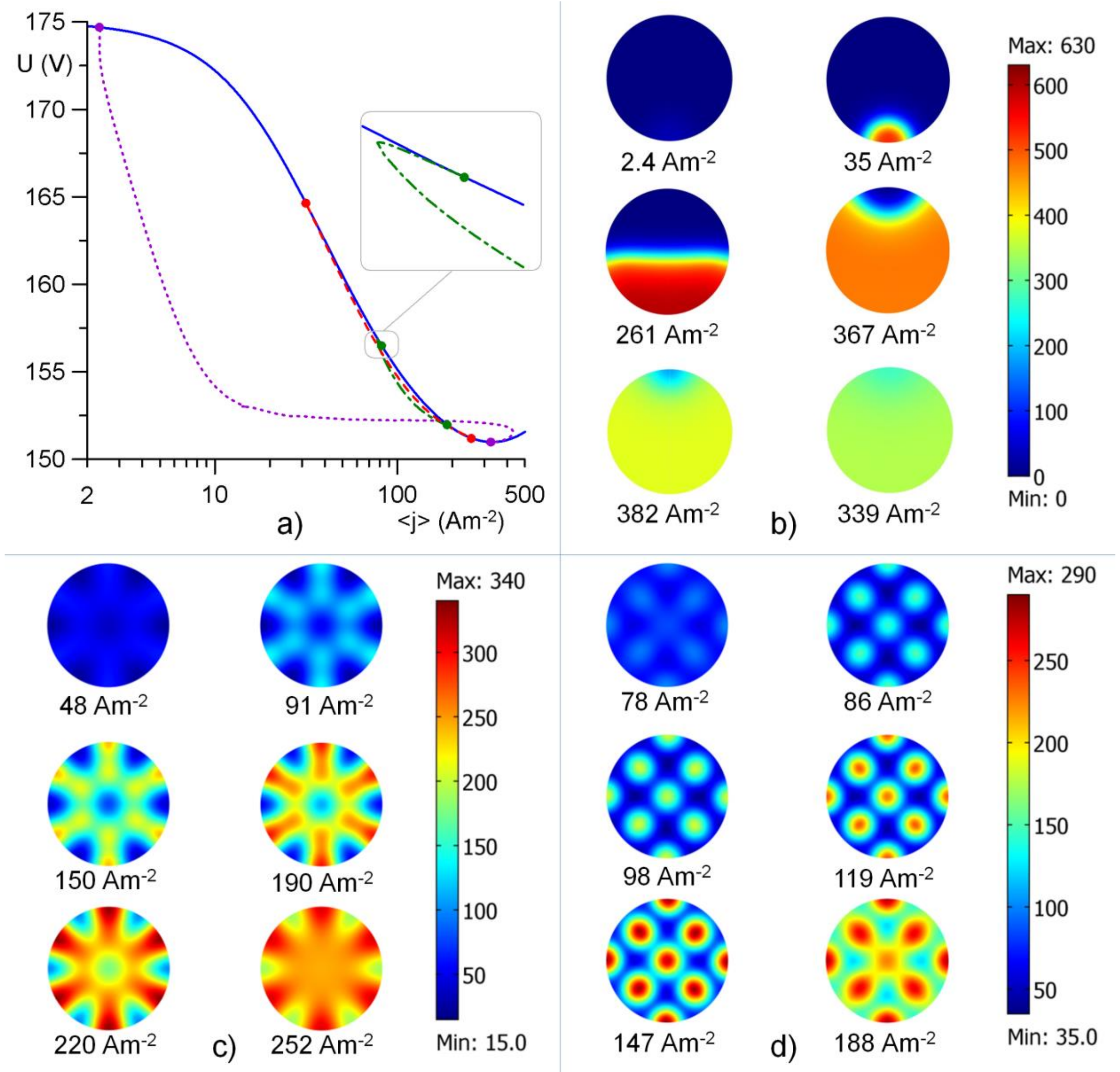
- Solid: the 1D mode.
- Dashed: the 1st 3D mode.
- Dashed-dotted: 8th 3D mode.
- Dotted: 12th 3D mode.

b): 1st 3D mode.

c): 8th 3D mode.

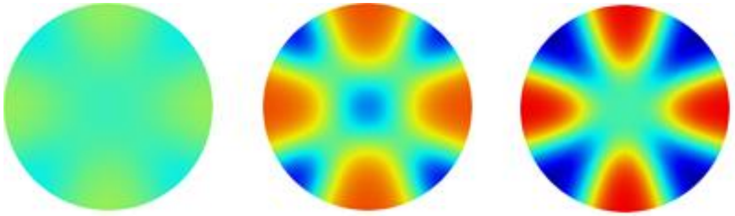
d): 12th 3D mode.

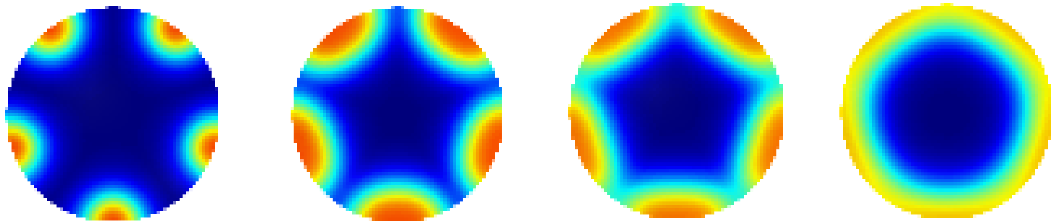
From Almeida *et al*, IEEE Trans. Plasma Sci. **39**, 2190 (2011).

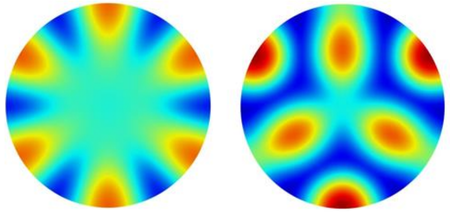


Self-organization on DC glow cathodes via stationary solvers

- There are quite a few observations of patterns in DC glow microdicharges.
- **The agreement between the modelling and the experiment is quite convincing**, although still qualitative up to now.

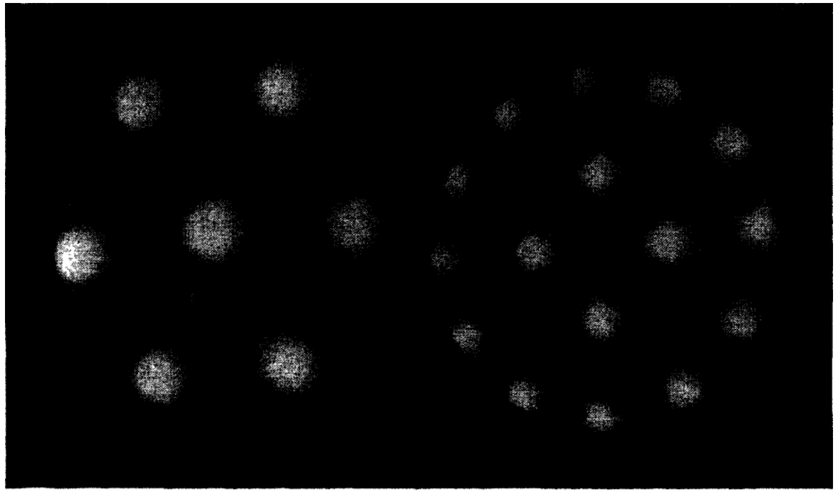




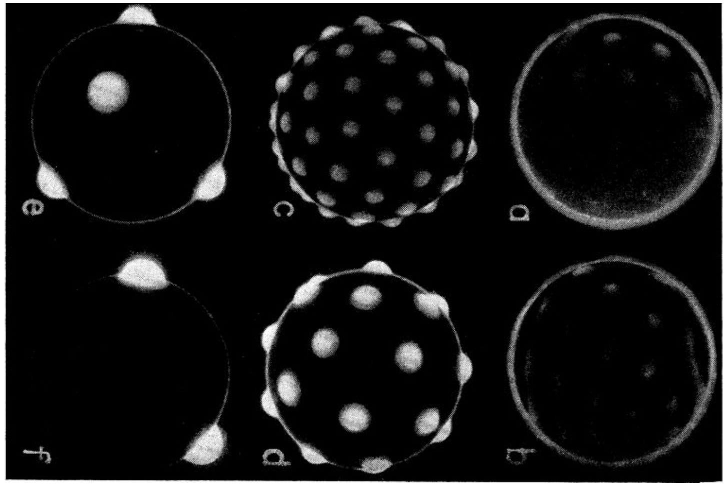


Bifurcations of steady states of glow discharge. Experiment: Zhu *et al*, Plasma Sources Sci. Technol. **23**, 054012 (2014). Modelling: Bieniek *et al*, Phys. Plasmas **25**, 042307 (2018).

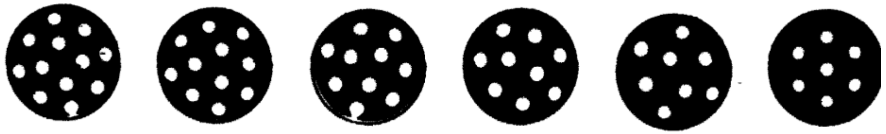
Self-organization on anodes of DC glow discharges

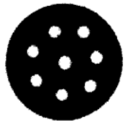


From Thomas and Duffendack, Phys. Rev. **35**, 72 (1930).



From Rubens and Henderson, Phys. Rev. **58**, 446 (1940).

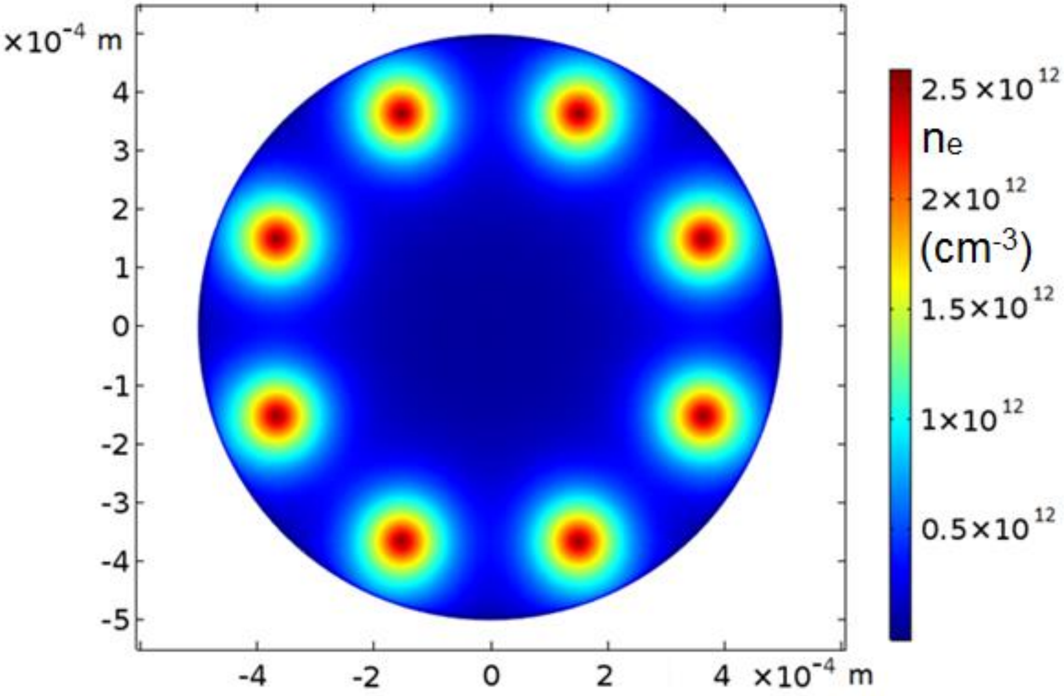




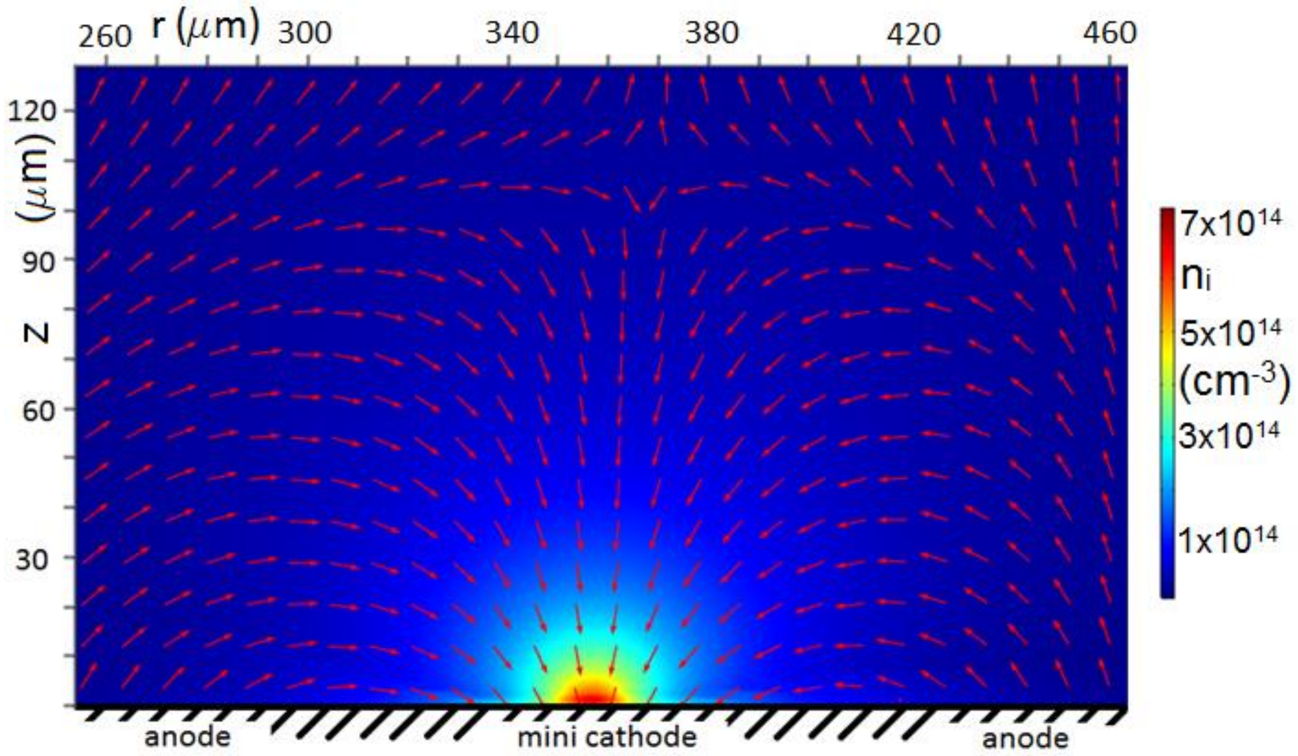
From Mueller, Phys. Rev. **A 37**, 4836 (1988).

From Arkhipenko *et al*, Plasma Sources Sci. Technol. **22**, 045003 (2013).

Self-organization on anodes of DC glow discharges via stationary solvers



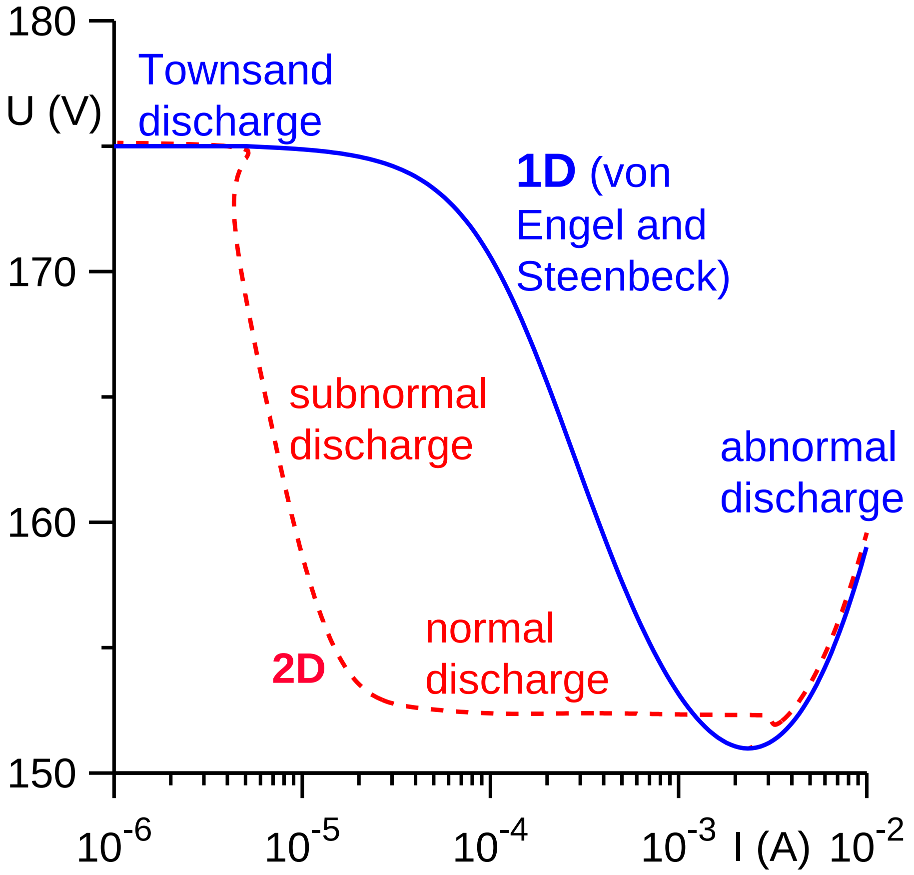
Electron density on the surface of the anode. I = 10 mA.

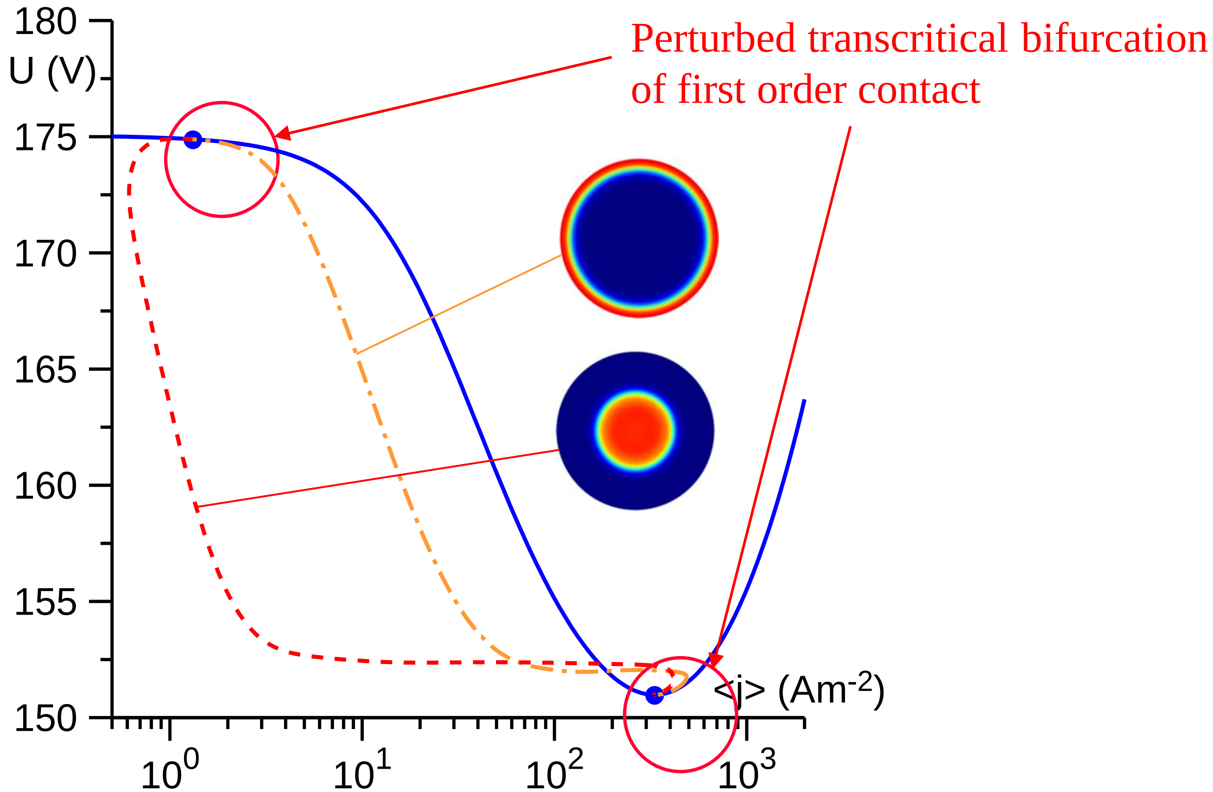


Ion density in the plane of symmetry passing through the spot center. Arrows: direction of the current density vector. I = 1 mA.

Self-organization on anode of discharge in He. 5 Torr, radius 0.5 mm, the height of the computation domain 5 mm. From Bieniek *et al*, Plasma Sources Sci. Technol. **27**, 05LT03 (2018).

Simple situations, complex behavior





How can diffusion of the charged particles to the wall, which is a weak effect (10^{-3}), originate such a huge difference?

Bifurcations may occur in apparently simple situations where multiple solutions are not of primary concern!

Glow discharge in Xe, 30 Torr, *R* = 1.5mm, *h* = 0.5mm. From Almeida *et al*, J. Phys. D: Appl. Phys. **42**, 194010 (2009).

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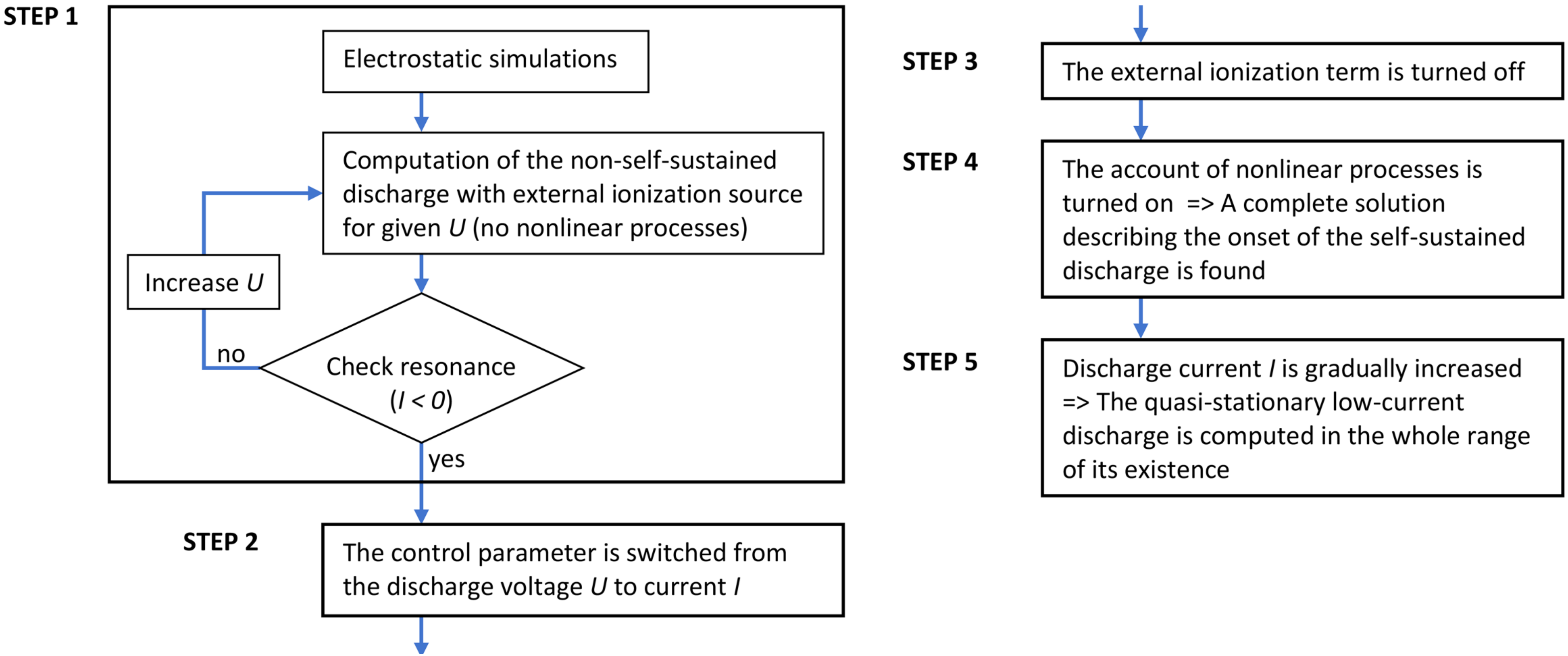
Modelling low-current quasi-stationary discharges by means of stationary solvers

- The most time-consuming step when using stationary solvers in nonlinear problems is finding a suitable initial approximation, which requires intelligent guesswork.

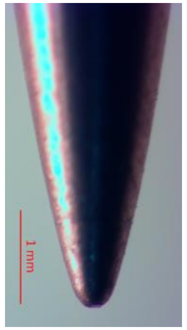
- The solution for each current serves as an initial approximation for simulations with the next current value. **But we need to start somewhere.**

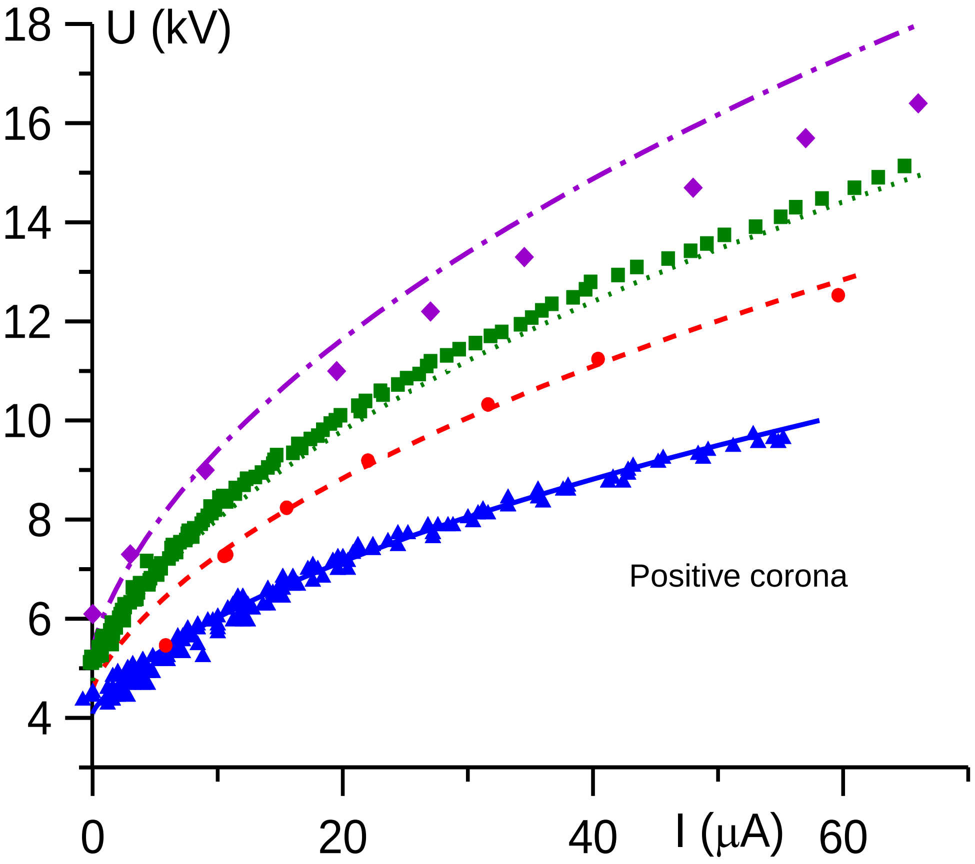
- **It is natural to start from the solution describing the initiation of the self-sustaining discharge**, provided by the resonance method.

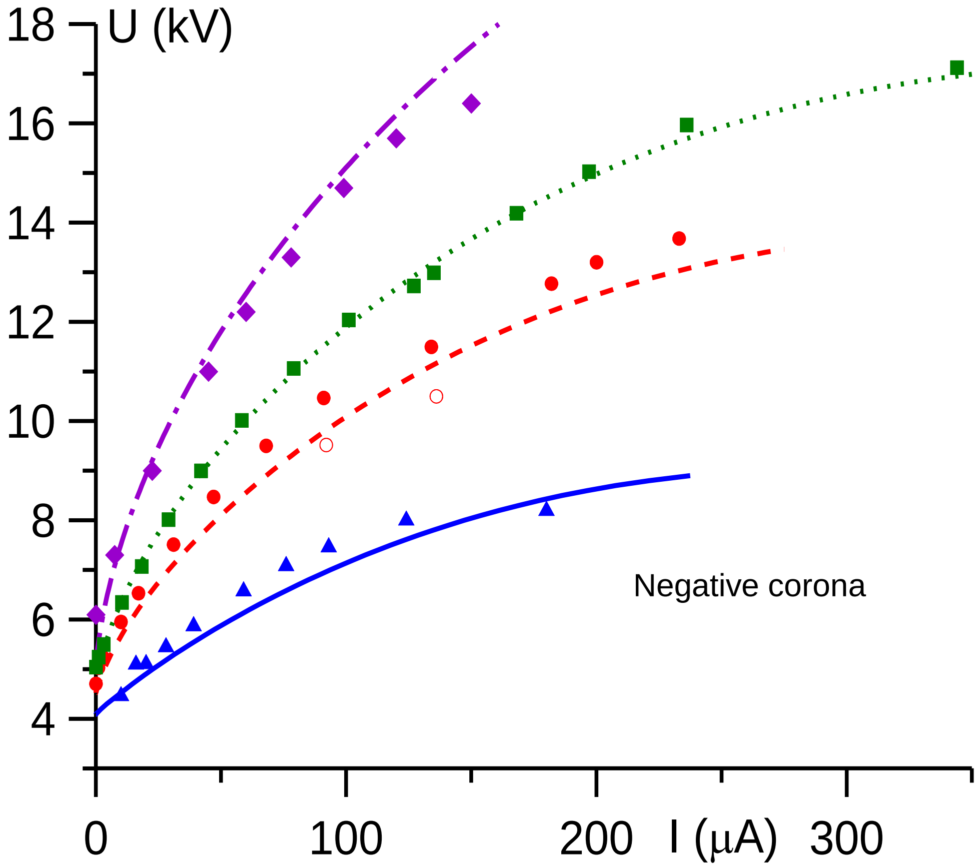
Integrated modelling of quasi-stationary low-current discharges



Integrated modelling of quasi-stationary low-current discharges







The CVCs given by the stationary solver are close to the time-averaged experimental CVCs for both positive corona, which is stable, and the negative corona, which is pulsed.

CVCs of point-to-plane coronas. Points: experimental time-averaged data. Lines: stationary modelling. From Ferreira *et al*, IEEE Trans. Plasma Sci. **48**, 4080 (2020).

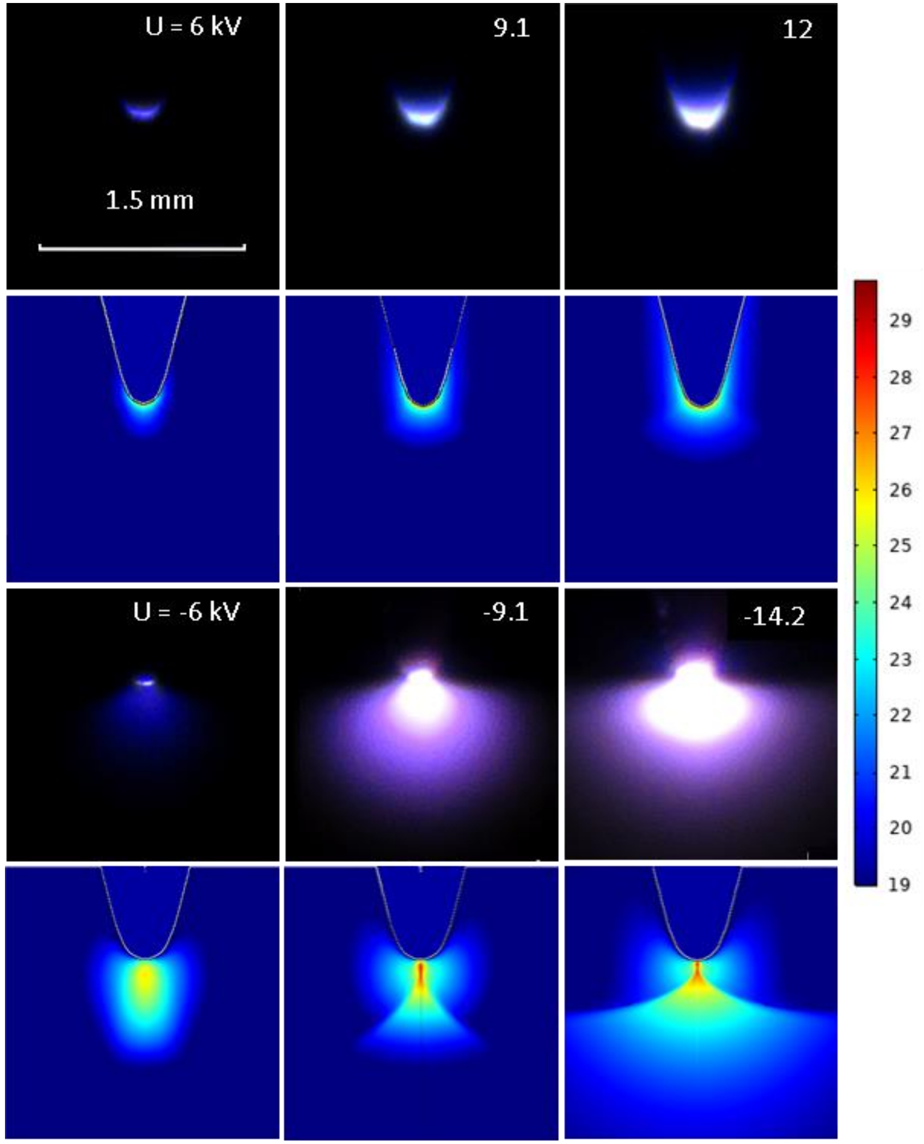
Triangles, solid: point-to-plane distance 5 mm. Circles, dashed: 8 mm.

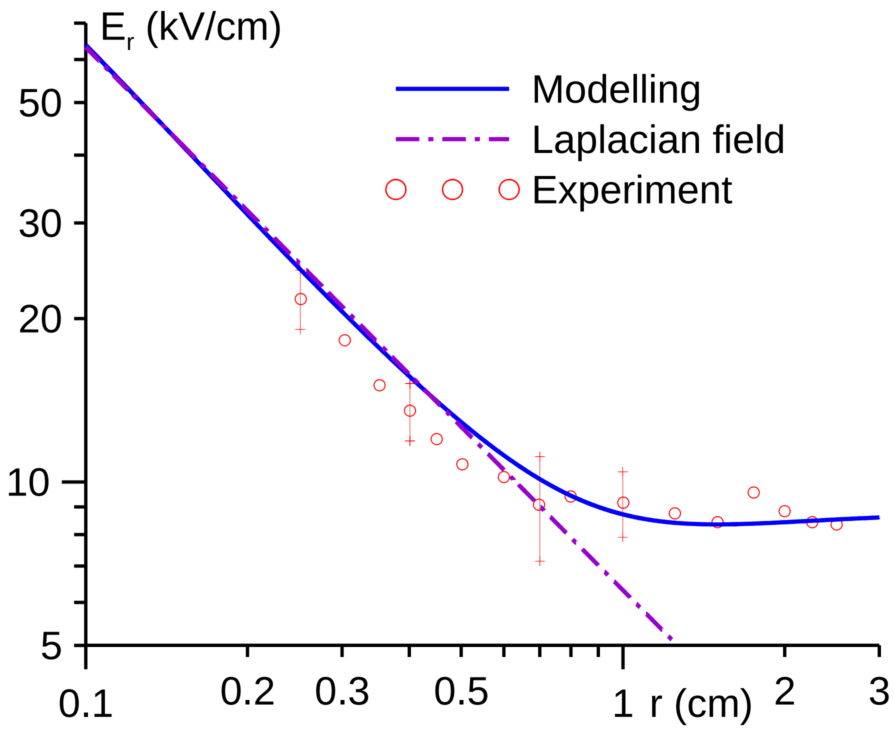
Diamonds, dash-dotted: 14 mm. Squares, dotted: 10 mm.

Integrated modelling of quasi-stationary low-current discharges

Images of point-to-plane corona discharges and computed distributions of the rate of excitation of radiating N_2. Gap 14 mm.

From Ferreira *et al*, IEEE Trans. Plasma Sci. **48**, 4080 (2020).





Electric field distribution in the negative corona discharge between concentric cylinders. Experiment: Cui *et al*, Appl. Phys. Lett. **115**, 244101 (2019).

From Ferreira *et al*, Appl. Phys. Lett. **117**, 026101 (2020).

Integrated modelling of quasi-stationary low-current discharges

Time of computation of solutions for U=5.05 and 5.1 kV. Initial approximation/initial condition: solution for U = 5 kV.

U (kV)	I (μA)	Stationary solver	Time-dependent solver
5.0	3.76	-	-
5.05	4.03	39s	58min
5.1	4.29	50s	5h 20min

Positive point-to-plane corona, gap 5 mm. From Ferreira *et al*, IEEE Trans. Plasma Sci. **48**, 4080 (2020).

- **Stationary solver is by orders of magnitude faster than time-dependent solvers.** Not surprising since the former is not subject to the Courant-Friedrichs-Lewy criterion or analogous limitations on time stepping.
- It was not possible to perform similar computations by means of the time-dependent solver for the negative corona, presumably because it is unstable => **Stationary solvers** allow decoupling of physical and numerical stability and **can be used for a fast direct calculation of time-averaged characteristics of pulsed discharges.**

Outline of the talk

- Introduction: low-current quasi-stationary gas discharges
- Model of low-current discharges in high-pressure air
- Computation of the initiation of self-sustaining gas discharges
- Modelling of quasi-stationary evolution of the discharge with increasing current
  - Stationary vs. time-dependent solvers
  - Integrated modelling of quasi-stationary low-current discharges
- **The determination of the current range where the quasi-stationary discharge becomes unstable**
- Conclusions

Stability of low-current quasi-stationary discharges

- Stationary solvers allow decoupling of physical and numerical stability => **It is possible to compute all the steady states of the discharge that are theoretically possible**, regardless of their stability and of whether they can be realized in a particular experiment with a given power supply.

- **After steady states of interest have been computed, their stability may be studied.**

- **Stability against small perturbations** may be studied by means of solving the eigenvalue problem resulting from the linear stability theory.

  Pro: - The eigenvalue problem governing stability of 2D (axially symmetric or planar) states against 3D perturbations, which are frequently the most dangerous ones, may be formulated and solved in 2D.

  Con: - Analysing a spectrum may be a laborious task.

- An alternative approach: to employ the same code that was used for calculation of the steady-state solution, **with the external circuit added and stationary solver replaced by a time-dependent one**.

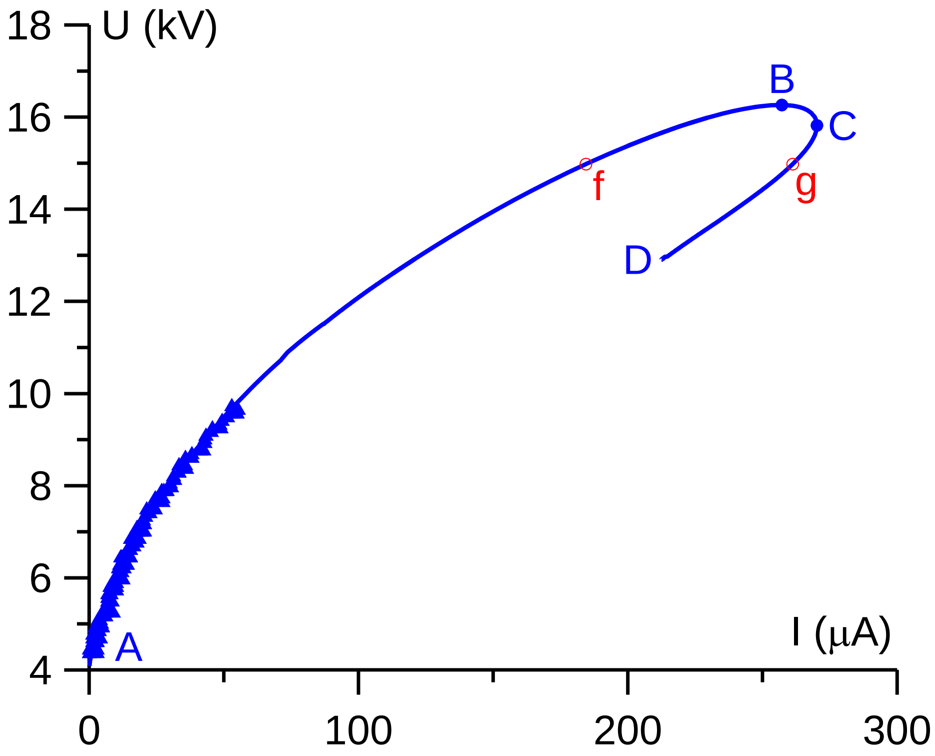
  Pros: - No need to solve an eigenvalue problem;

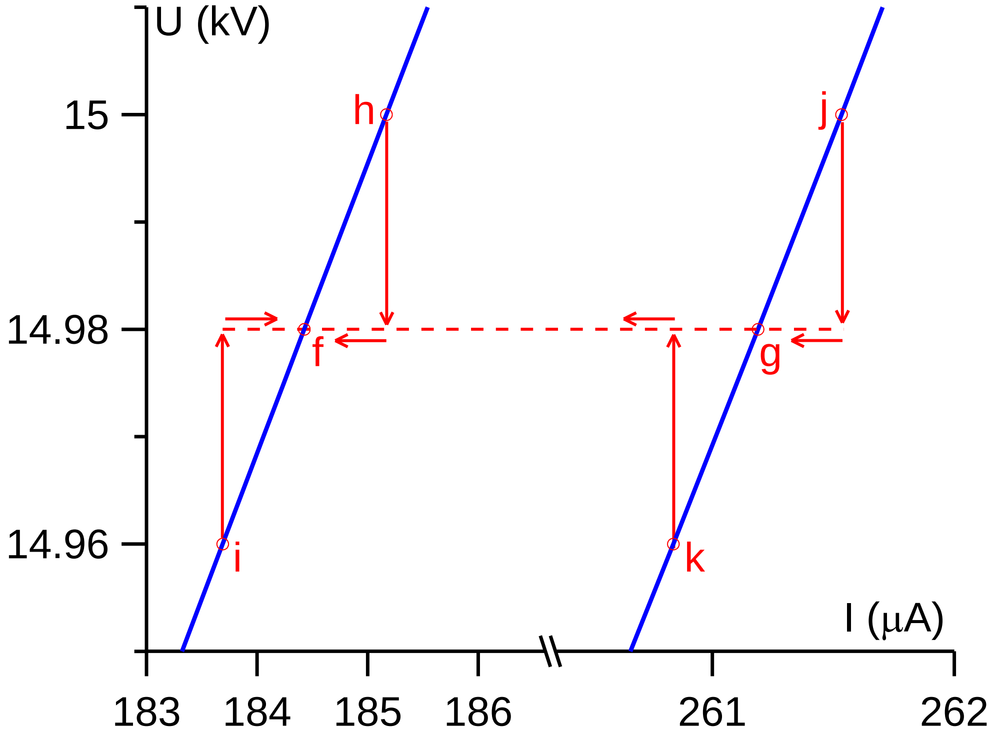
  - Stability against both small and finite perturbations.

  Cons: - In certain cases a time-dependent solver computes some unstable steady states and/or is unable to compute stable states [Almeida *et al*, Plasma Processes Polym. **14**, 1600122 (2017)];

  - Time-dependent solvers give no information on stability against perturbations of symmetries different from the one of the calculation domain of the solver. For instance, 2D time-dependent simulations say nothing about stability against 3D perturbations.

Example: stability of positive corona





CVC of a positive point-to-plane corona discharge in a wide current range. Air, 1 atm, gap 5 mm. From Benilov *et al*, J. Appl. Phys. **130**, 121101 (2021).

- **Steady states on the ascending branch of the CVC are stable against perturbations of a limited amplitude.** The limit is ~150 V for lower *U* and decreases as *U* increases. Larger perturbations evolve into streamers.
- **States after the maximum are unstable against all perturbations.** The discharge either relaxes to the steady state with the same *U* on the ascending branch on the CVC, or no stationary state is reached and streamers are formed.

Conclusions

- **The ignition of self-sustained gas discharges is governed by a multidimensional boundary-value eigenvalue problem for** a system of stationary PDEs.
- The eigenvalue problem results in the classical Townsend self-sustainment criterion, provided that similar simplifications are applied (parallel-plate geometry, single ion species, no diffusion, no photoionization).
- **The physical sense of the eigenvalue problem is similar to that of the Townsend criterion**: the applied voltage should be just sufficient for the direct ionization by electron impact and photoionization to compensate losses of the charged particles.
- Three different methods for its solution have been analyzed. In the authors' experience, **the resonance method is the method of choice**.
- **The method is based on solving linear PDEs and can be implemented with the use of standard solvers, including commercial ones.** It is physics-based, fast, and has been tested in a wide range of conditions and complex geometries.

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Conclusions

- **Modelling of quasi-stationary evolution of the discharge with increasing current may be conveniently performed using stationary solvers**, starting from the inception-point solution computed by the resonance method.
- **Stationary solvers are faster than time-dependent solvers**, in many cases by orders on magnitude, and offer other important advantages.
- **Stationary solvers can compute all the steady states of the discharge that are theoretically possible**, regardless of their stability.
- **After steady states of interest have been computed, their stability may be studied**, either by means of solving the eigenvalue problem resulting from the linear stability theory, or by means of replacing the stationary solver by a time-dependent one.

Thank You!

Thank You for your patience and your attention!

More detail:

Journal of Applied Physics

A practical guide to modeling low-current quasi-stationary gas discharges: Eigenvalue, stationary, and time-dependent solvers

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