Plasma Physics 2





Lecture series contents

- 1. Townsend breakdown theory, Paschen's law
- 2. Glow discharge
- 3. Electric arc at low and high pressures
- 4. Magnetized low-pressure plasmas and their role in material deposition methods.
- 5. Brief introduction to high-frequency discharges
- 6. Streamer breakdown theory, corona discharge, spark discharge
- 7. Barrier discharges
- 8. Leader discharge mechanism, ionization and discharges in planetary atmospherres
- 9. Discharges in liquids, complex and quantum plasmas
- 10. Thermonuclear fusion, Lawson criterion, magnetic confinement systems, plasma heating and intertial confinement fusion.

Where are we in the IV chart?

- We have seen last time that the deformation of the E field in various real-life scenarios leads to a formation of non-uniform discharge structures (streamers, arcs, coronas).
- Today, we are expanding further on those by analyzing DBDs which operate in Townsend/Glow part of the IV characteristic



volt-ampérová charakteristika elektrických DC výbojů (vznik jednoho mikro-výboje bariérového výboje můžeme pro daný čas uvažovat jako DC)

Barrier discharge fundamentals

- A barrier discharge is characterized by the fact that there is a dielectric between the electrodes, preventing the formation of a conductive channel an electrical short cannot occur
- Some fundamental configs (Brandenburg 2017):



• Often filamentary, here is a glow discharge at the atm. pressure:



General barrier discharge mechanism

- (Dielectric) barrier discharge occurs between electrodes, at least one of which is covered by a dielectric.
- An important driver for DBD formation is charge accumulation on the dielectric.
- When described by an equivalent circuit, it behaves like a capacitor.
- Due to the dielectric, it always has to be driven by an AC field



Dielectric barrier discharge – ancient history

• Ancient history - G.C.Lichtenberg (1777):





a) Vid. Scripta Academiae Suec. Scientiarum ad ann. 1762.



Brief history of the dielectric barrier discharge



• Ozone generation, Lichtenberg figures, streak measurements.



W. Siemens 1857 and Buss 1932, Klemenc 1937, Manley 1943, Samoilovich 1966, Gibalov 1981, Eliasson, Kogelschatz 1983, Heuser 1985, Okazaki 1993, Zhu, Hidaka 1996, Guikema 2000, Kozlov 2001, Wagner 2010

Initial observations of DBD discharges

• Optical and electrical studies historically performed in Aachen (Pietsch, Heuser, Gibalov) and in ABB (Hirth, Kogelschatz, Eliasson):



Lichtenberg figures



General forms of the DBD discharge

• Basic modes of the DBD are filamentary and (Brandenburg 2004):



- In the filamentary mode, the fialment diameter is sub-millimeter. Individual filaments také nanoseconds and the electric field is hundreds of Td owing to the streamer mechanism.
- In the diffuse mode, the discharge size is given by electrode width but the mode is not universaly stable.
- Penning ionization one of phenomena contributing to the stability of the discharge
- Diffuseness is important for various applications of the discharge. Note that even the filaments can be distributed uniformly and the discharge is pseudo-diffuse.

Diffusion barrier discharges (Massines)



Diffusion barrier discharge in He

Diffusion barrier discharge in N2



CURRENT IN DBD DISCHARGES

Measuring electrical current in a DBD discharge

- You can either measure current using induction-based probes or by measuring voltage on a parallel resistor.
- Equivalent circuit for a single-filamentary discharge in air.







Synek et al. 2018 PSST

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Electrical characterization of a DBD discharge

• To measure the actual current is challenging, because the Maxwell translation current makes a major contribution.



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Analyzing electrical measurements

• Single-filamentary volume barrier discharge in atmospheric pressure argon.



Figure 2. Supply voltage, cell current, and 5- μ s time-integrated photographs of the discharge, in the visible range for $P_{\rm Ar} = 400$ Torr, f = 10 kHz, $\boldsymbol{Q} = 2$ mm, $U_{\rm max} = 2.36$ kV.





Analyzing electrical measurements

You cannot simply subtract the sine component to get the correct current

- 3 К 2 1.0 V(t)2 HV Q(t)= 6.5 kHz f 1.1 0.5 C_d 1 $U = 12 \text{ kV}_{pp}$ 1 1 Voltage u_{cell} (kV) 1 1 i_{cell} (mA) ١. Metal. -1 1 1 d = 1.2 mm -0.5 1.1 -2 R = 7.5 mm 1 1 $\Delta = 1.5 \text{ mm}$ 1.1 2 mr -3 -i(t)0 20 Range of 1.1 -1.0 1 1 Scanning Ŧ d 1 1 Figure 2. Supply voltage 60 Current ١. photographs of the discha ١. Glass $P_{\rm Ar} = 400$ Torr, f = 101. 1 1.1 $= \frac{1}{1 - \frac{C_{cell}}{C_t}} \left[i(t) - C_{cell} \frac{\mathrm{d}V(t)}{\mathrm{d}t} \right]$ 1.1 4000 1 1 3000 1 1 1 1 $\mathsf{R}_{\mathsf{meas}}$ 2000 т т **T** 0001- voltage 2π 0 $\varphi_i \varphi_{i+1}$ urrent [A] enlarged 0.0 in part b) -2000 $j_g(t) = C_g \frac{\mathrm{d}U_g(t)}{\mathrm{d}t}$ -0.5 -3000 -0.10 -4000 -1.0 90 180 270 360 0 -0.5 0.0 0.5 1.0 1.5 2.0 a) phase [°] t (µs)
- Single-filamentary volume barrier discharge in atmospheric pressure argon.

An alternative method for obtaining j_R







From Kirchhoff's laws

$$j_R(t) = \frac{1}{1 - \frac{C_{cell}}{C_d}} \left[i(t) - C_{cell} \frac{\mathrm{d}V(t)}{\mathrm{d}t} \right]$$

Conduction current Translational
current
$$i_t(t) = i_c(x, t) + \varepsilon(x)\varepsilon_0 \frac{\partial E(x, t)}{\partial t} = i(t)/A$$
Measured
 $i_c(x, t) = q[\Gamma_p(x, t) - \Gamma_e(x, t)]$

$$\stackrel{i_t(t)}{=} \int_0^{d+g} \frac{dx}{\varepsilon(x)} = \int_0^{d+g} \frac{i_c(x, t)}{\varepsilon(x)\varepsilon_0} dx + \frac{\partial}{\partial t} \int_0^{d+g} E(x, t) dx$$
 $j_t(t) \left(\frac{1}{C_d} + \frac{1}{C_g}\right) = \frac{1}{A\varepsilon_0} \int_0^{d+g} \frac{j_c(x, t)}{\varepsilon(x)} dx + \frac{dV(t)}{dt}$
 $\varepsilon(x \le g) = 1$
 $j_t(t) \left(\frac{1}{C_d} + \frac{1}{C_g}\right) = \frac{1}{C_g} j_c(t) + \frac{dV(t)}{dt}$
 $j_R(t) = j_c(t) = \frac{1}{1 - \frac{C_{cell}}{C_d}} \left[i(t) - C_{cell} \frac{dV(t)}{dt}\right]$

An alternative method for obtaining j_R

• If we take into account the deposited surface charge, then (Bonaventura 2020):

$$i_t(t) = i_c(x,t) + \varepsilon(x) \frac{\partial E_g(t)}{\partial t} = \varepsilon(x)\varepsilon_0 \frac{\partial E_d(t)}{\partial t} = i(t)/A$$

surface charge deposition

step condition on the boundary of a dielectric (Tirpák)

 $\frac{\partial \sigma}{\partial t} = i_c \qquad \qquad E_g = \varepsilon_r E_d - \sigma/\varepsilon_0$

after a few modifications:

$$\frac{j_t(t)}{A} = \int_0^g j_c(x,t) dx + \left(\frac{\partial \sigma}{\partial t}\right) \left(\frac{dg(\varepsilon_r - 1)}{\varepsilon_r g + d}\right) - \frac{g + d}{\varepsilon_r g + d} \varepsilon_r \varepsilon_0 \frac{\partial V}{\partial t}$$
$$= \frac{\partial \sigma}{\partial t} \left[\frac{\varepsilon_r g(g + d)}{\varepsilon_r g + d}\right] + \frac{g + d}{\varepsilon_r g + d} \varepsilon_r \varepsilon_0 \frac{\partial V}{\partial t}$$
$$\downarrow$$
$$j_R(t) = j_c(t) = \frac{1}{1 - \frac{C_{cell}}{C_d}} \left[i(t) - C_{cell} \frac{dV(t)}{dt}\right]$$

the calculated current j_R obtained from Kirchhoff laws is identical to conduction current j_C

QV diagrams of the DBD discharge

- Manley 1943 showed that the energy dissipated in a DBD can be obtained from Lissajouse figures, graphically.
- Today, these plots are called more accurately QV plots/diagrams



Figure 2. Supply voltage, cell current, and 5- μ s time-integrated photographs of the discharge, in the visible range for $P_{\rm Ar} = 400$ Torr, f = 1 **g** Hz, d = 2 mm, $U_{\rm max} = 2.36$ kV.



QV diagrams of the DBD discharge



• Power dissipated in the discharge can be obtained from current and voltage:

 $P(t) = j_R(t)U_g(t)$

• And for total dissipated energy, it has to hold that:

$$\int_0^T i(\tau) V(\tau) \mathrm{d}\tau = \int_0^T j_R(\tau) U_g(\tau) \mathrm{d}\tau$$

VOLUME DIAGNOSTICS OF DBD PLASMAS

Applying cross-correlation spectroscopy to understand DBDs (Kozlov 2001)







200

10 4

10 5

Intensity scale:

Number of counts:

300

10²

10³

400

10¹

Time (ns)

1

Light emission during the discharge

Measuring the electric field in DBDs

• Spectroscopic method based on nitrogen emission bends, developed by Kozlov in 2001

$$SPS$$

$$e + N_{2}(X^{1}\Sigma_{g}^{+})_{\nu=0} \rightarrow N_{2}(C^{3}\Pi_{u})_{\nu'=0} + e$$

$$(\Delta E = 11.0 \text{ eV}),$$

$$N_{2}(C^{3}\Pi_{u})_{\nu'=0} \rightarrow N_{2}(B^{3}\Pi_{g})_{\nu''=0} + h\nu$$

$$(\lambda_{C} = 337.1 \text{ nm}),$$

$$N_{2}(C^{3}\Pi_{u})_{\nu'=0} + N_{2}/O_{2} \xrightarrow{K_{N_{2}}^{C}/K_{O_{2}}^{C}} \text{ Products},$$

$$\frac{dn_{C}(r, t)}{dt} = k_{C} \left(\frac{E}{n}\right) n_{N_{2}}n_{e}(r, t) - \frac{n_{C}(r, t)}{\tau_{eff}^{C}}$$

$$\frac{1}{\tau_{eff}^{C}} = K_{N_{2}}^{C}n_{N_{2}} + K_{O_{2}}^{C}n_{O_{2}} + \frac{1}{\tau_{0}^{C}}$$

$$= K_{N_{2}}^{C}n_{N_{2}} + K_{O_{2}}^{C}n_{O_{2}} + \sum_{\nu''=0}^{\infty} \frac{1}{\tau_{0\nu''}^{C}}$$





Obrusnik et al. 2005 JPD Obrusnik et a. 2018 PSST Bilek et al. 2018 PSST



Spatio-temporal evolution of DBDs



Spatially resolved simulations of DBD discharges

• Simulating a co-planar DBD discharge, Jánský 2022



Figure 6. Two-dimensional side-view development of the discharge electric field strength (logarithmic scale), electron (logarithmic scale) and surface charge densities for coplanar discharge in atmospheric pressure air. Case for photoemission of 5×10^{-5} .

SURFACE CHARGE IN DBD DISCHARGES

Surface charge in DBDs

Pockels effect has been used to measure surface charge (Zhu 1996), for a point-to-plane config

Positive tip transitioning to negative polarity

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Negative tip transitioning to positive polarity



Surface charge in DBDs

- There is a "surface streamer" forming in point-to-plane DBDs (Kumada 2009)
- The electric field exceeded an equivalent of 30kV/cm at higher pressure (experiments done at low pressure)





Figure 14. Computed results. Distribution of (a) potential V; (b) horizontal component of electric field, $\sqrt{E_x^2 + E_y^2}$; (c) perpendicular component of electric field, E_z ; (d) intensity of electric field, |E|.

SELF ORGANIZATION OF FILAMENTARY DBD DISCHARGES

Self organization of filamentary DBDs (Guikema 2000)

Localized filaments – they "prefer" to ignite in discrete places, rather than randomly.

The decisive mechanism is the surface charge accumulation and the "dose" of charge deposited by individual discharges.







střední vzdálenost filamentů



Self organization of filamentary DBDs

Microscopic view of the charge accuulation effect





DBD APPLICATIONS

Ozone generation (Eliasson 1987)



- Possibly oldest application of DBD discharges
- Motivated formulation of the first physical and chemical models.
- 1D model described the formation of a transient glow discharge at high pressure.
- Actuall EEDF was calculated and from it, rate coefficients and transport properties were deduced.

The mechanism for ozone formation and potential ozone loss was identified as follows:

$$e + O_2 \rightarrow 2O + e$$

 $O + O_2 + M \rightarrow O_3 + M$

 $O + O_3 \rightarrow 2O_2$

Ozone production – oxygen dissociation



Figure 6. Simplified potential energy diagram of O_2 . The arrow indicates the Franck–Condon region for excitation from the ground state.



Figure 7. Distribution of electron energy losses as a function of the reduced electric field; energy branching in oxygen.

$$e + O_2 \rightarrow e + O_2 (A \ {}^{3}\Sigma_{u}^{+})$$

$$\rightarrow e + O ({}^{3}P) + O ({}^{3}P)$$

$$e + O_2 \rightarrow e + O_2 (B \ {}^{3}\Sigma_{u}^{+})$$

$$\rightarrow e + O ({}^{3}P) + O ({}^{1}D)$$

$$O ({}^{1}D) + O_3 \rightarrow 2O_2$$

Classically, the Franck-Condon principle is the approximation that an electronic transition is most likely to occur without changes in the positions of the nuclei in the molecular entity and its environment. The resulting state is called a Franck-Condon state, and the transition involved, a vertical transition. The quantum mechanical formulation of this principle is that the intensity of a vibronic transition is proportional to the square of the overlap integral between the vibrational wavefunctions of the two states that are involved in the transition.



Ozone production – effect of admixtures

In industrial production, gases that form metastable excited species are often added, further enhancing the dissociation rate of oxygen by plasma.

$$e + N_2 \xrightarrow{k_{17}(E/n)} N_2^* + e$$

 $N_2^* + O_2 \longrightarrow N_2 + O + O$



Figure 6-32. Influence of admixtures of N₂ and CO on ozone yield in a DBD in room-temperature oxygen: black circles correspond to molecular nitrogen, white circles correspond to carbon monoxide.



Ozone production – an industrial setup





Surface processing

SOFTAL GmbH





pro reaktivní plyny a depozice (Massines 2012)



změna kontaktního úhlu, PET povrch, DCSBD (Homola 2017)

DCSBD – in-line opracování, no-pinholing, i tepelně citlivé povrchy/vzorky





Excimer light sources

$R^* + R \rightarrow R_2^*$ $R_2^* \rightarrow 2R + hv$					R^* - excited atom R_2^* - excited dimer						(AB* = exciplex)			
Ar [*] ₂ Kr [*] ₂	F *	Xe [*]	ArCl*	ArF*	KrCl*	KrF*	XeI*	Cl ₂ *	XeBr*	Br [*] 2	XeCl*	I_2^*	XeF*	
126 146	157	172	175	193	222	248	253	259	282	289	308	342	354	



Very intense UV light sources

- surface and water cleaning by UV
- UV curing and photopolymerization
- excimer lasers for semiconductor applications

Excimer light sources









Very intense UV light sources

- surface and water cleaning by UV
- UV curing and photopolymerization
- excimer lasers for semiconductor applications

Takeaways

- What is a DBD and what are its various arrangements
- Where is the voltage drop in DBDs, what are the methods for accurately measuring plasma power and voltage?
- What are some methods of DBD diagnostics.
- DBD applications