Online Low Temperature Plasma Seminar
6th September, 2022

https://theory.pppl.gov/news/seminars.php?scid=17&n=oltp-seminar-series

Cathodic arc plasma science and applications: The known unknown and unknown known

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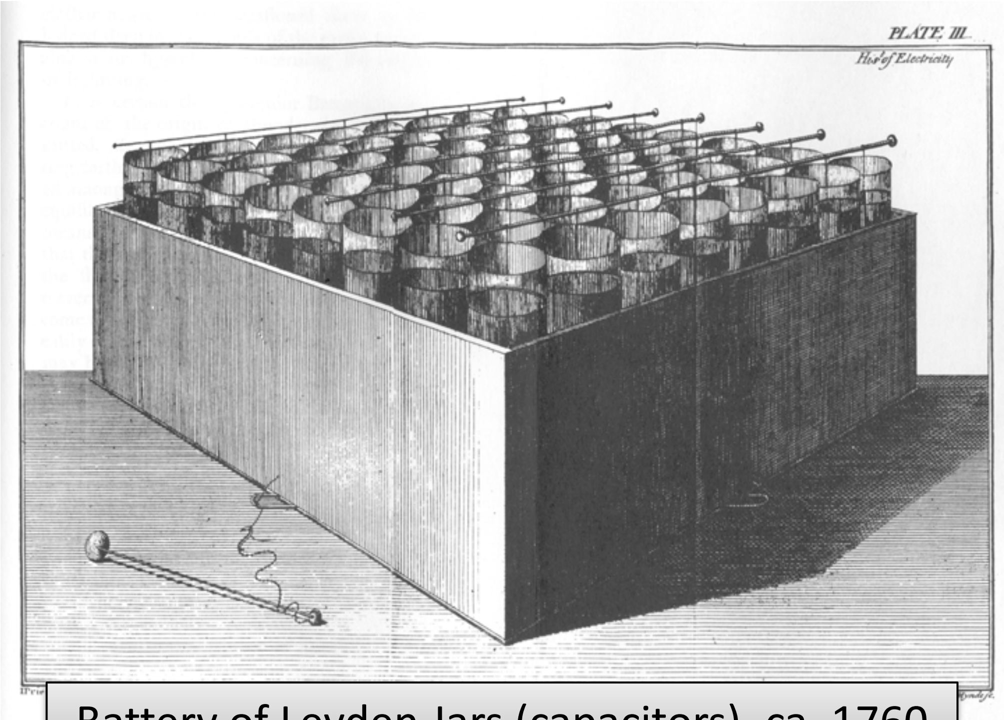
Colleagues, past and presents, in Berkeley, Leipzig and beyond, whose contributions are mentioned, are gratefully acknowledged!

IOM

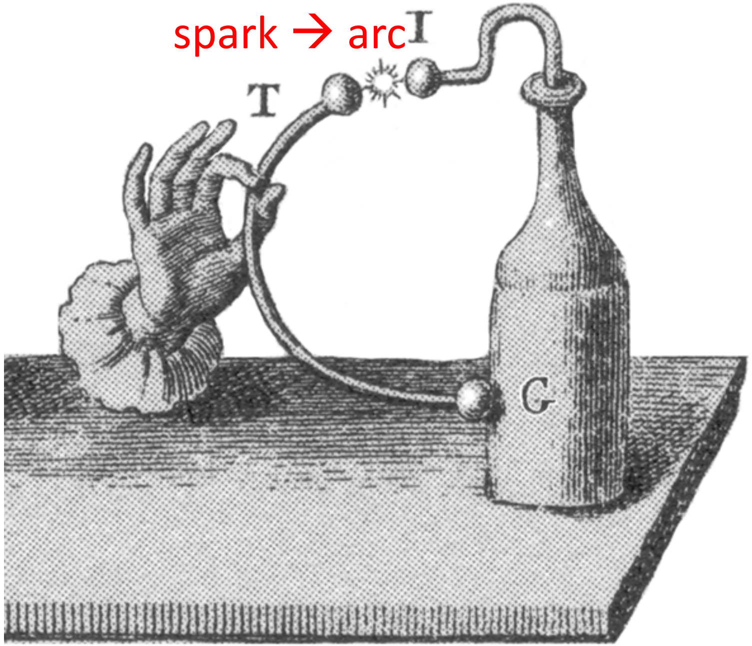
Cathodic arc plasma science and applications: The known unknown and unknown known

→ to be on solid ground, focus on arc plasma physics:
there are many (unknown) knowns and (known) unknowns …

Pulsed Arcs in Air, (unintentional) Energetic Deposition



Battery of Leyden Jars (capacitors), ca. 1760



Alessandro Volta, ca. 1765

J. Priestley, The History and Present State of Electricity, London 1766

A. Anders, *IEEE Trans. Plasma Sci*. 31, 1052 (2003), and “Cathodic Arcs”, Springer, NY 2008.

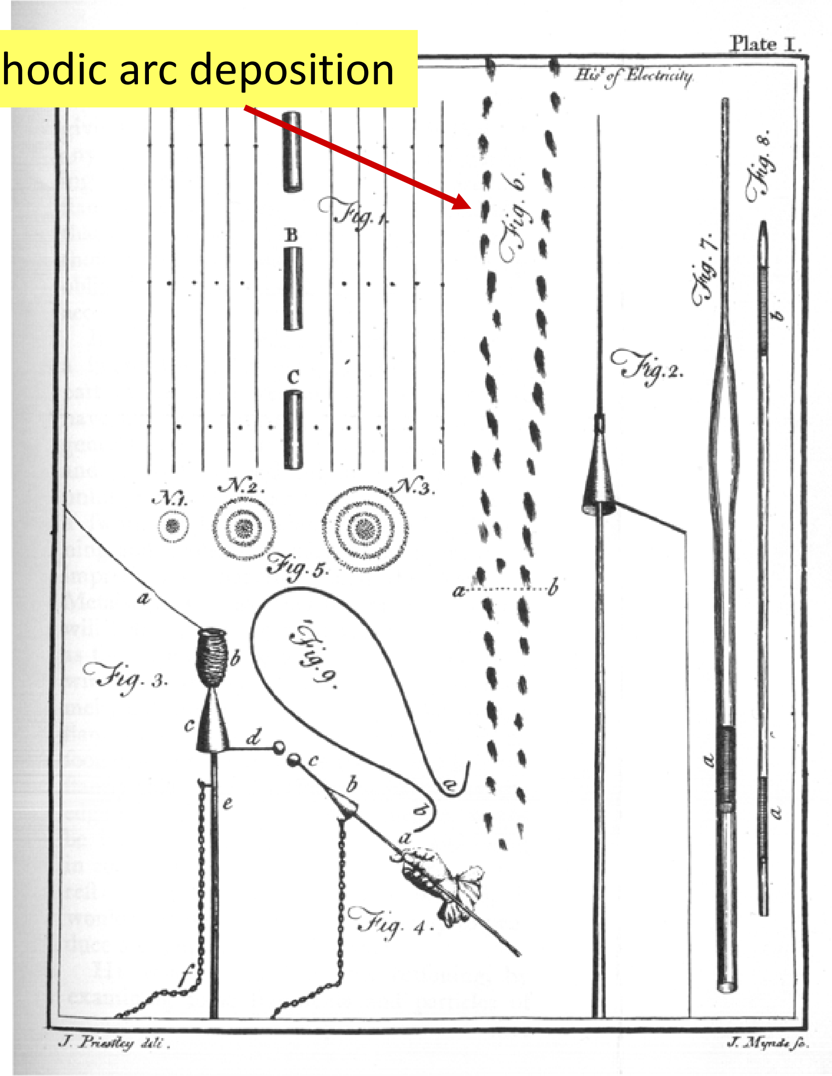
1766 - Joseph Priestley

- Cathodic arc coating (in air)
- discharge of a bank of Leyden jars through a brass chain
- arcs between each link of the chain
- deposit on glass is well adherent
- observed Newton's rings (interference, oxide films)
- found black coating (copper oxide)

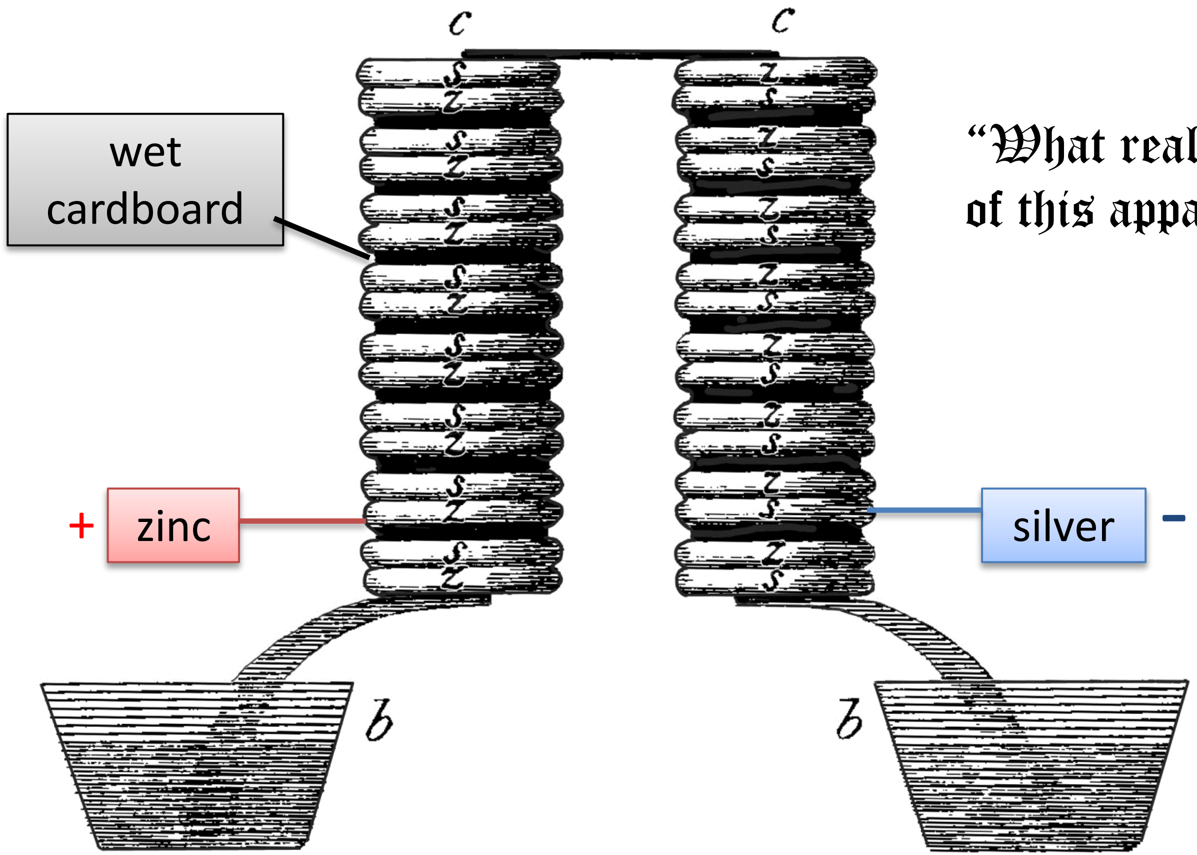
J. Priestley, The History and Present State of Electricity, London 1766

A. Anders, *IEEE Trans. Plasma Sci*. 31, 1052 (2003), and "Cathodic Arcs", Springer, NY 2008.

Cathodic arc deposition



Volta, 1800: The "Pile" – A New Electrical Energy Source



"What really increases the electric power of this apparatus is the number of plates"
Volta, 1800

A. Volta, Phil. Mag. VIII (1800) 289.

1802/03 Vasilii Petrov

- First continuous arcs in air and at low pressure using an “enormous battery” of 4200 copper-zinc plates
- published only in Russian, his work was unknown or ignored

(on arc history) A. Anders, IEEE Trans. Plasma Sci. 31 (2003) 1052

ИЗВѢСТІЕ

о

ГАЛЬВАНИ - ВОЛЬТОВСКИХЪ

ОПЫТАХЪ,

которые производилъ

Профессоръ Физики Василій Петровъ,

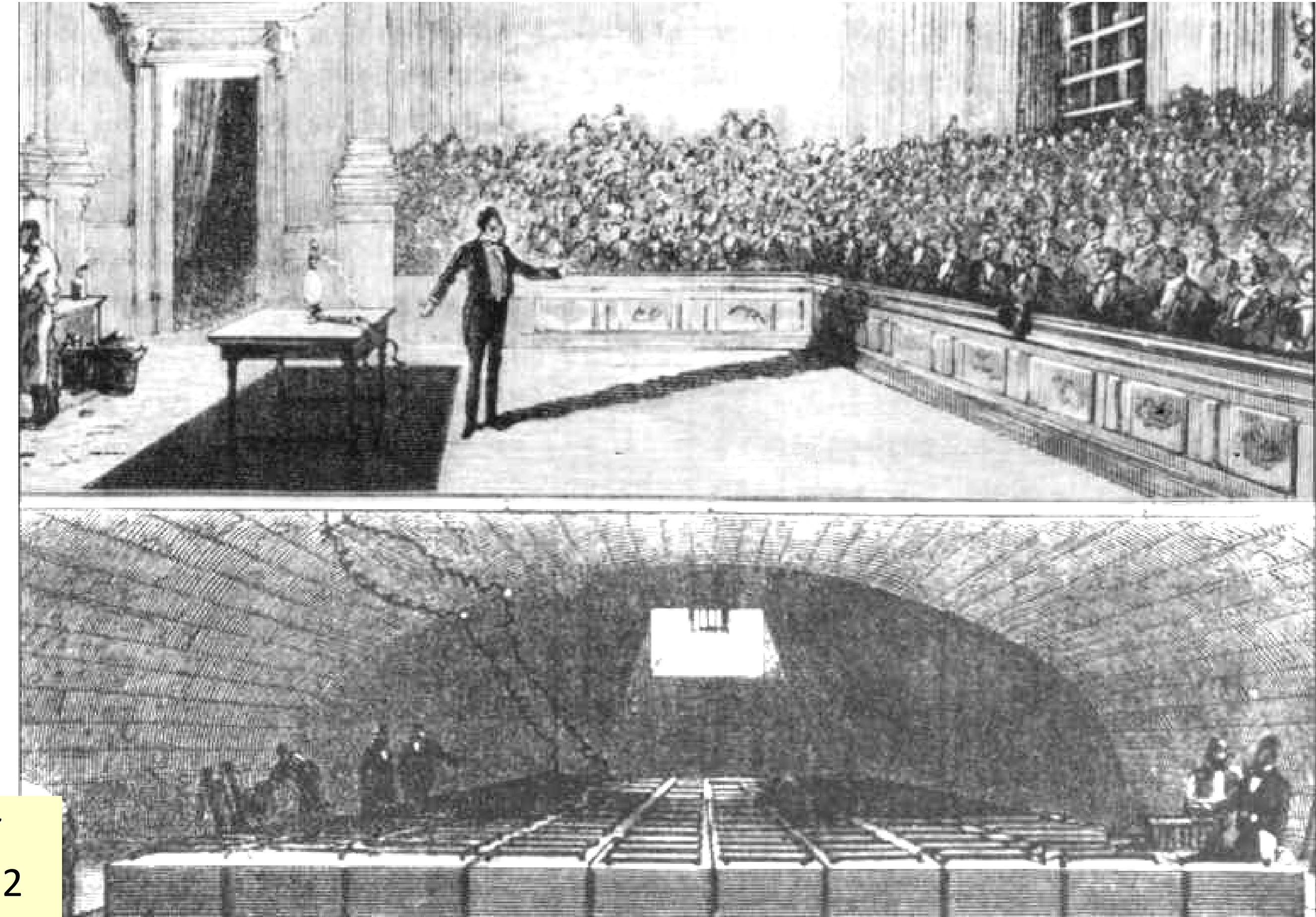
посредствомъ огромной наипаче баттереи, состоявшей *иногда* изъ 4200 мѣдныхъ и цинковыхъ кружковъ, и находящейся при Санкт-Петербургской Медико-Хирургической Академіи.

ВЪ САНКТ-ПЕТЕРБУРГѢ,

Въ Типографіи Государственной Медицинской Коллегіи, 1803 года.

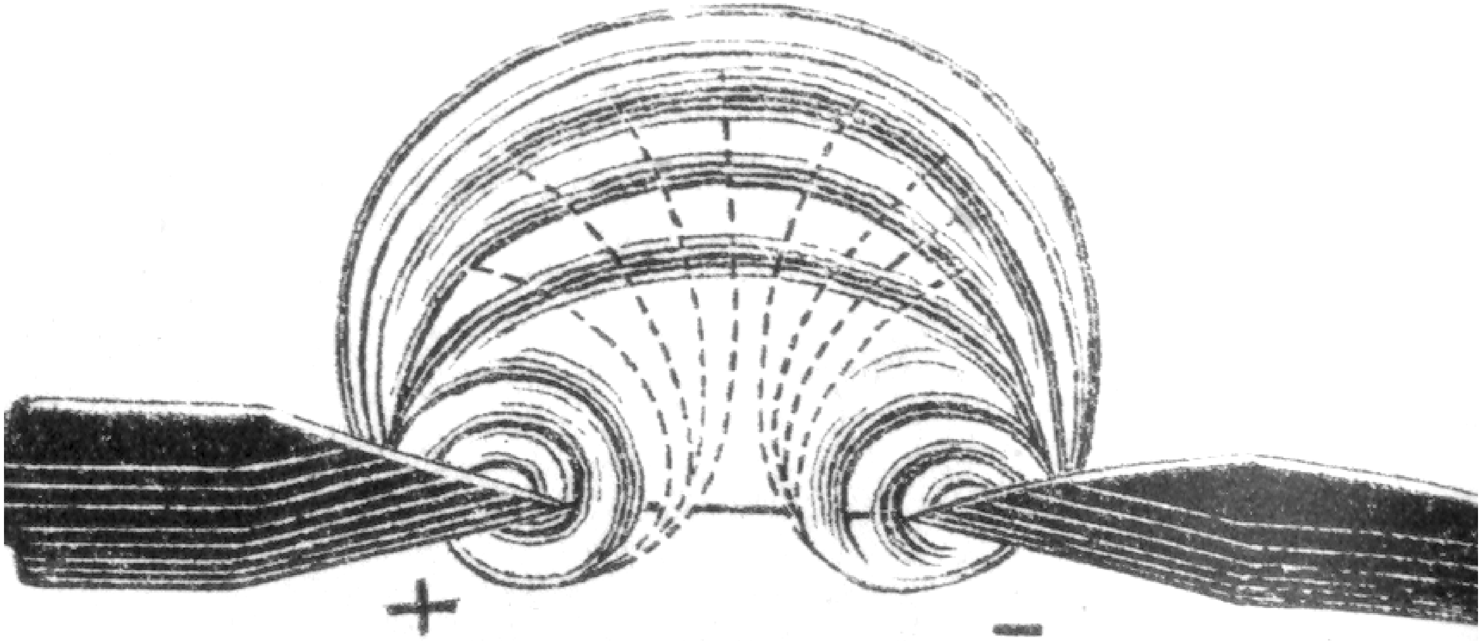
1802…1821 – Arc Discharge Demonstrations

- Humphry Davy at the Royal Institution London: Continuous arcs in air and in low-pressure vessels (1809)



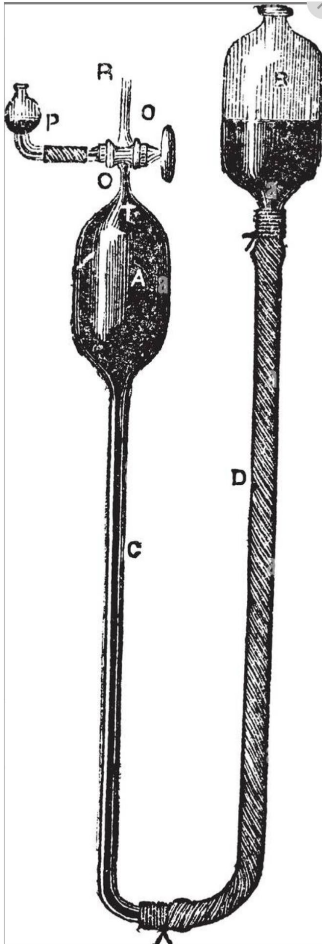
(on arc history) A. Anders, *IEEE Trans. Plasma Sci*. 31 (2003) 1052

Arc Discharge

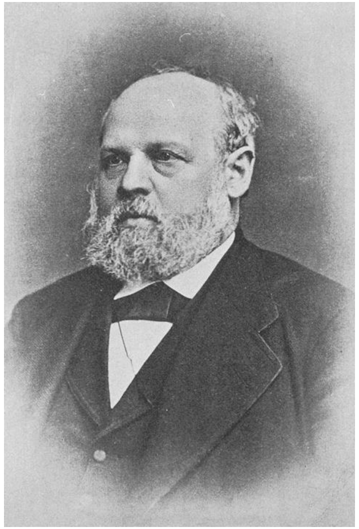


G. Wiedemann, Die Lehre von der Elektricität, Braunschweig 1885

Important: Improved vacuum pumps of the 19th centrutry



- Geissler‘s idea (1855) to use mercury as a „piston“ to pump air: then the base pressure is given by mercury‘s vapor pressure (0.5 Pa at room temp.)
- used by Edison to pump his light bulbs

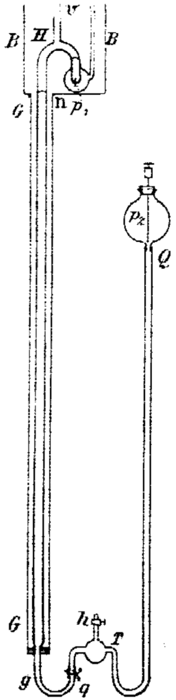


Heinrich Geissler (1814–1879)

Mercury Arc

Leo Arons (1892)

"Despite the continual impression of the appearance of light, the discharge is always discontinuous; the discontinuity is easily recognized by a telephone receiver, which is connected next to the apparatus in addition to a large capacitor. The discontinuity is due to the fact that, despite the measured low voltage at the electrodes, an electromotoric force almost three times as large is required to produce the arc; 50 volts are just sufficient."



Jahrg. 11. Nr. 6.

Verhandlungen

der

Physikalischen Gesellschaft zu Berlin.

Sitzung vom 21. October 1892.

Vorsitzender: Hr. A. Kundt.

Hr. **W. Jäger** berichtete auf Grund von gemeinsam mit Hrn. **D. Kreichgauer** angestellten Versuchen

Ueber den Temperaturcoefficienten des Quecksilbers.

Hr. **L. Arons** sprach dann

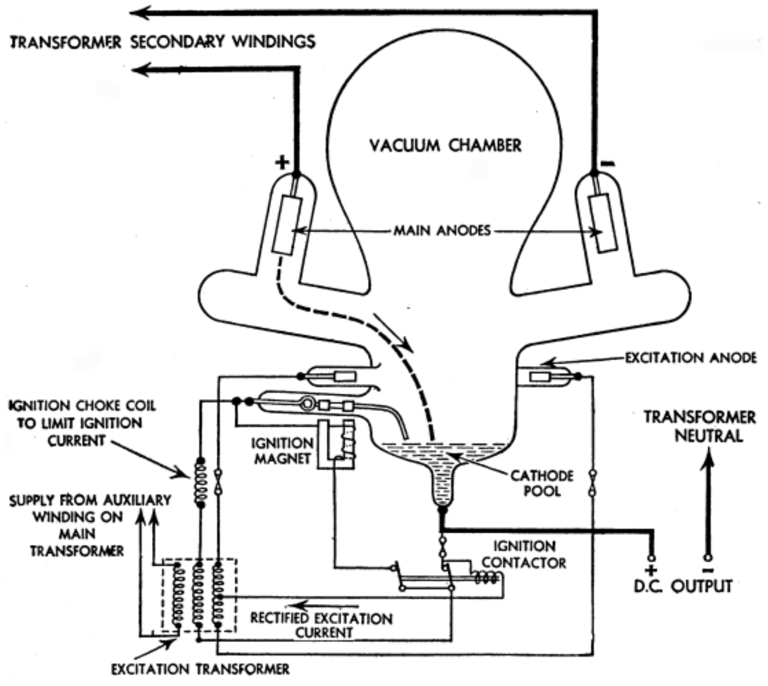
Ueber einen Quecksilberlichtbogen.

Mit Versuchen über die Gasentladung beschäftigt, fand ich eine ausserordentlich einfache Methode, einen intensiv leuchtenden, lang andauernden Lichtbogen zwischen Quecksilberelectroden herzustellen, welcher nur verhältnissmässig geringe electromotorische Kräfte zu seiner Unterhaltung erfordert und keine der lästigen Eigenschaften von Lichtbögen zwischen Metallelectroden besitzt.

L. Arons, Verhandlungen der Physikalischen Gesellschaft zu Berlin, 11 (1892) 767-771.
L. Arons, (Wiedemann's) Annalen der Physik und Chemie, 58 (1896) 73-95.

Mercury arc rectifiers

also known as *Cooper-Hewitt* or *Hewittic rectifiers* were extensively used to provide DC in high power applications, powers ranging from kW to MW, with voltages from 110 V to 30 kV.



http://www.electricstuff.co.uk/mercarc.html

Arc rectifier used in a NY substation for 90 years

Cathode Spots

Fig. 5.

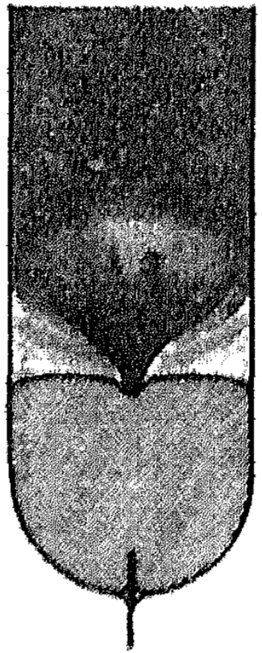
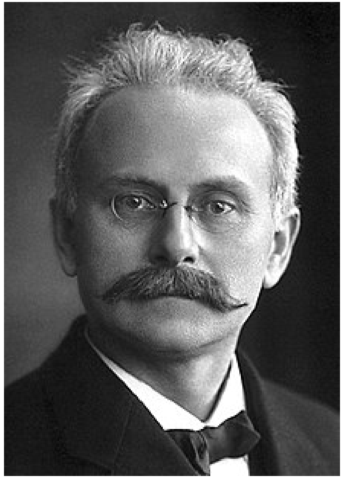


Fig. 6.

Fig. 7.



Johannes Stark (1874-1957)
(same person who found broadening of spectral lines due to electric fields)

Arc spot on mercury cathode , displacement when magnetic field is present

J. Stark, Zeitschrift für Physik, 4 (1903) 440-443.

Cathode Spots:

Q1: Why do they form?
Q2: Why are they non-stationary?

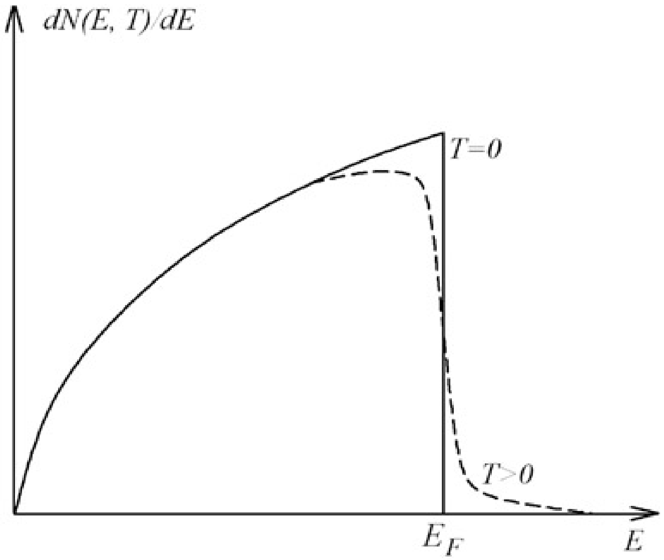
A1: Because thermionic and field emission mechanisms are nonlinear with surface temperature and electric field

A2: Because they change the emission environment in nonlinear ways

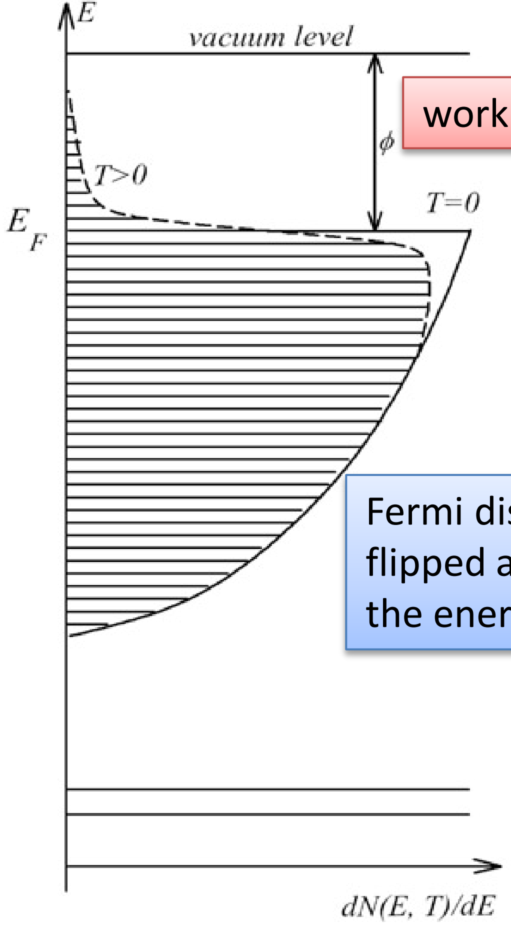
A. Anders, Cathodic Arcs, Springer, NY, 2008

Electrons in Metals: Fermi Distribution

Fermi-Dirac distribution function



$$\frac{dN(E,T)}{dE} = \frac{4\pi(2m_e)^{3/2}}{h^3} \frac{E^{1/2}}{1+\exp\left(\frac{E-E_F}{kT}\right)}$$

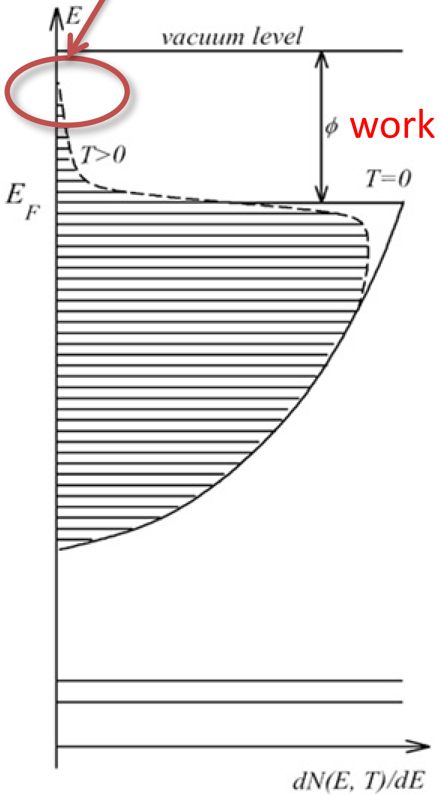


work function

Fermi distribution function flipped and rotated as to have the energy axis pointing up

Thermionic Emission from Hot Metal

those hot electrons, the tail of the distribution, can go OVER the potential barrier



Richardson-Dushman equation

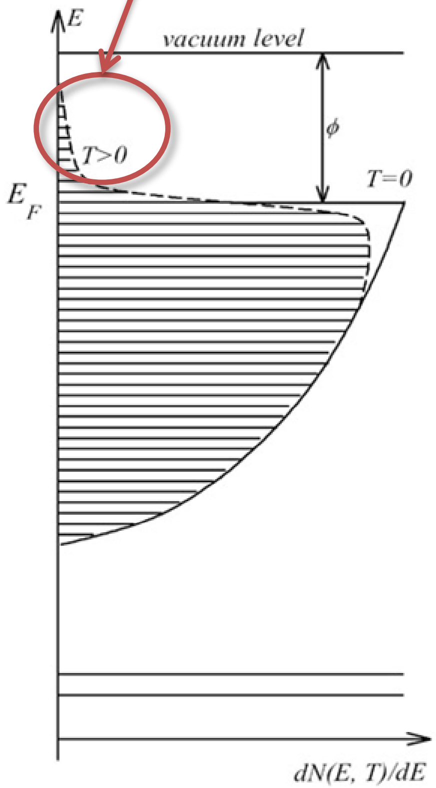
$$\boxed{j_{thermionic} = AT^2 \exp\left(-\frac{\phi}{kT}\right)}$$

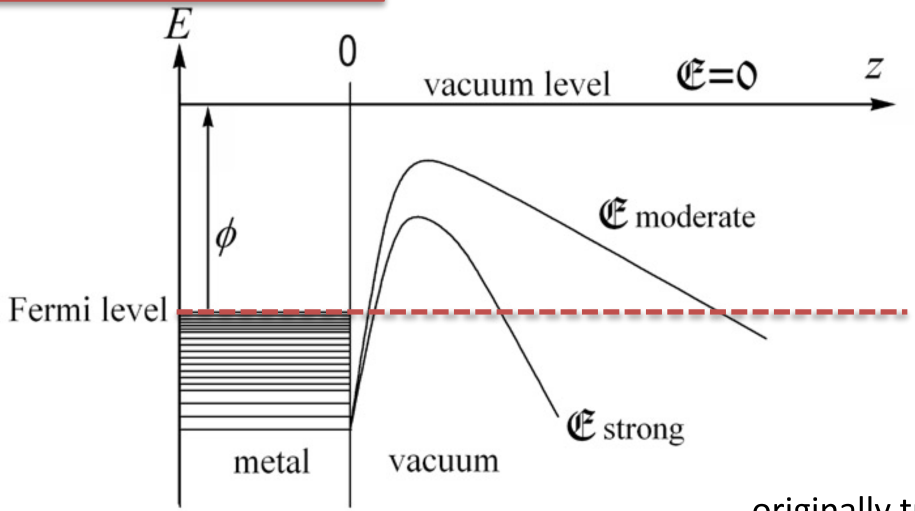
with the Richardson constant

$$A = \frac{4\pi e m_e k^2}{h^3} = 1.202\times10^6 \ \mathrm{A/m^2K^2}$$

Field Emission of Electrons

less energetic electrons can tunnel THROUGH the potential barrier, if **E**-field is strong





reduction of work function

$$\phi_S = \phi - \left(\frac{e^3 \mathfrak{E}}{4\pi\varepsilon_0}\right)^{1/2}$$

originally treated by Fowler-Nordheim
(is rather complicated, even for T = 0)

$$\boxed{j_{FN}(\mathfrak{E}) = \frac{e^3}{8\pi h\,\phi\, t^2(y)} \mathfrak{E}^2 \exp\left[-\frac{8\pi\sqrt{2m_e\phi^3}}{3eh\mathfrak{E}} v(y)\right]}$$

$$y \equiv \sqrt{\frac{e^3\mathfrak{E}}{4\pi\varepsilon_0}\,\frac{1}{\phi}}$$

elliptical functions

Thermo-Field Emission of Electrons

Combining classical "**over** the barrier" with quantum-mechanical "**through** the barrier" gives a somewhat complicated expression for the emission current density:

$$j_{TF}\left(T,\mathfrak{E},\phi\right)=\frac{4\pi m_e kT}{h^3}\left\{\int_{-W_A}^{W_l}\frac{\ln\left[1+\exp\left(-\frac{E_z+\phi}{kT}\right)\right]}{1+\exp\left[\frac{8\pi\left(2m_e\right)^{1/2}v\left(y\right)E_z^{3/2}}{3he}\right]}dE_z+\right.$$
$$\left.+\int_{W_l}^{\infty}\ln\left[1+\exp\left(-\frac{E_z+\phi}{kT}\right)dE_z\right]\right\}$$

The rest of the definition of all symbols would take 2 more slides….
they can be found in chapter 3 of

A. Anders, Cathodic Arcs: From Fractal Spots to Energetic Condensation, Springer, New York, 2008.

Thermofield-Emission to Explosive Emission

Dynamics of cathode spots: carbon arc as seen with a high-speed framing camera
arc current up to 5 kA, 200 ns exposure time per frame

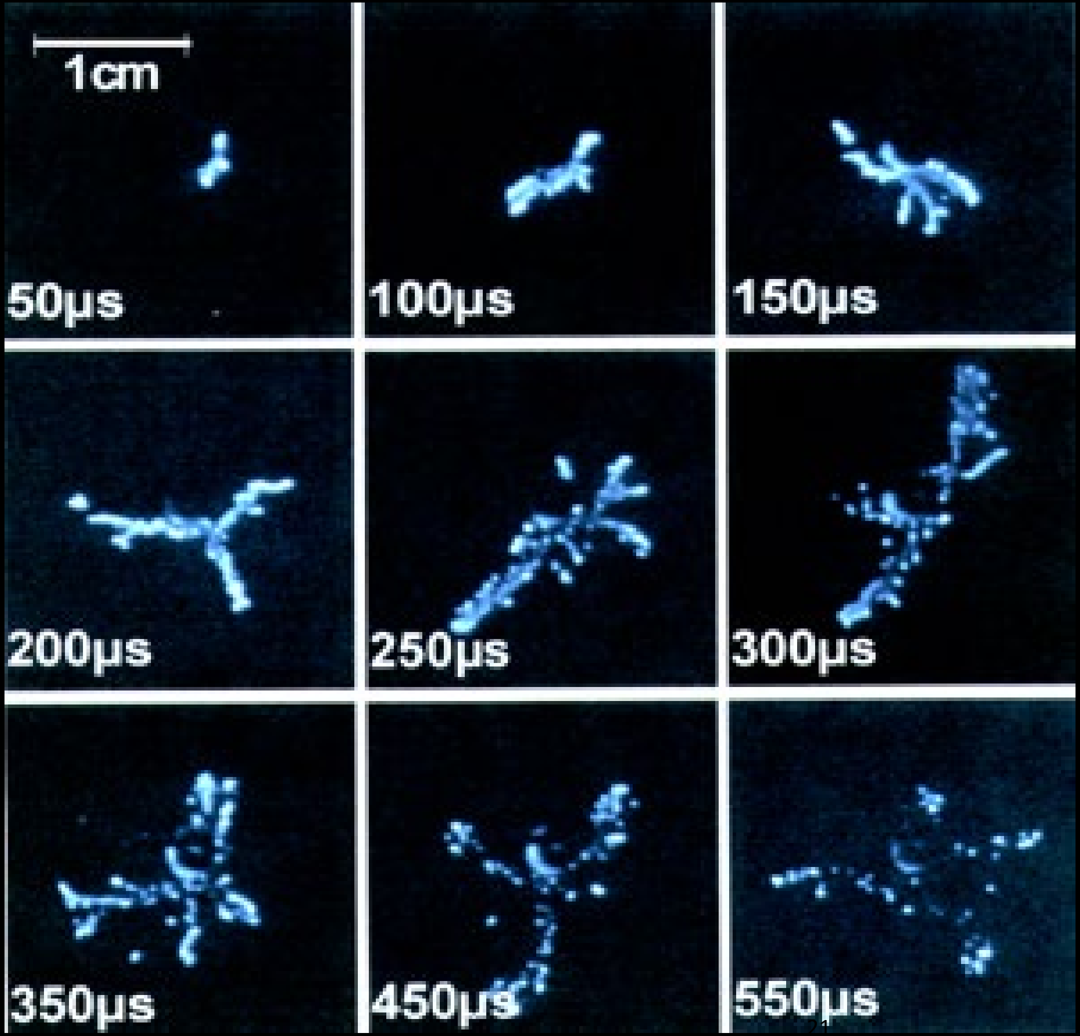
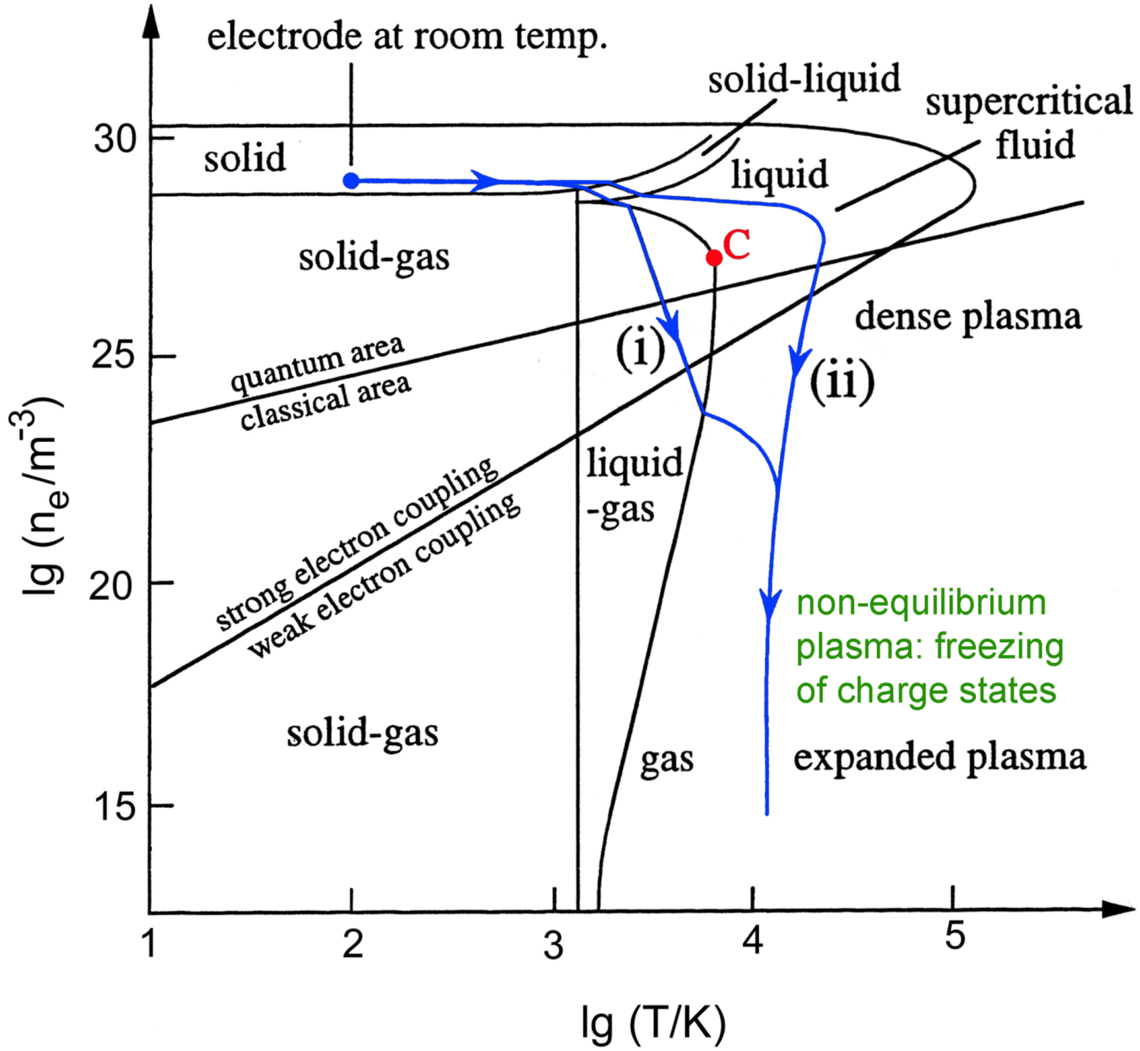


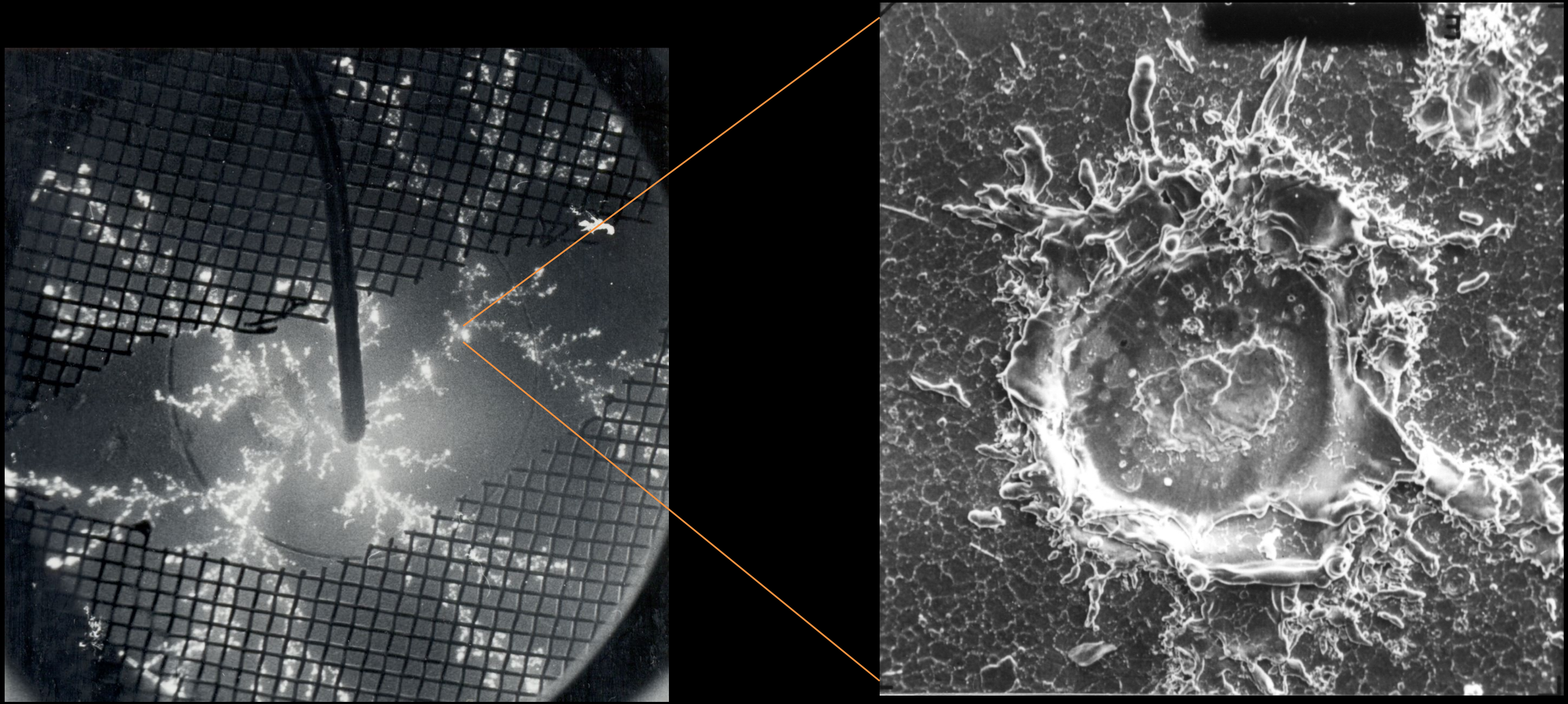
Figure: Courtesy of P. Siemroth and T. Schülke (about 2000).

Path of the Cathode Material in the *n*-*T* Phase Diagram



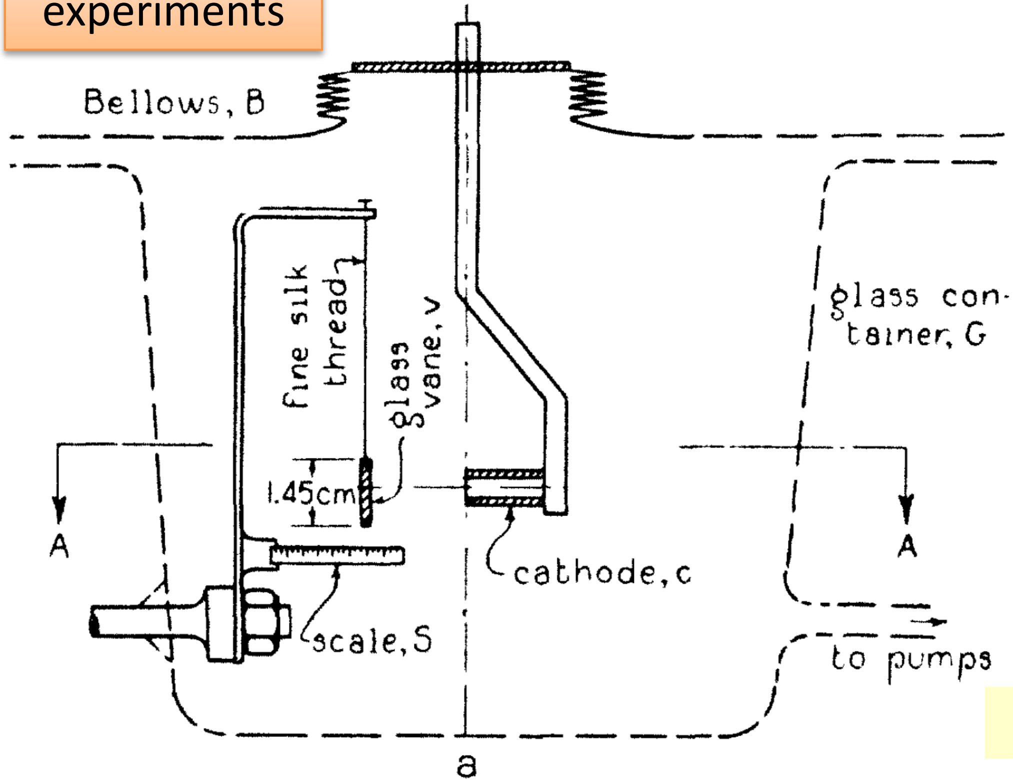
A. Anders, Plasma Sources Sci. Technol. **21** (2012) 035014.

Effects of high power density in cathode spot



Supersonic ion flow

Pendelum experiments



→ Cu ion velocity = 1.6 x 10^4 m/s

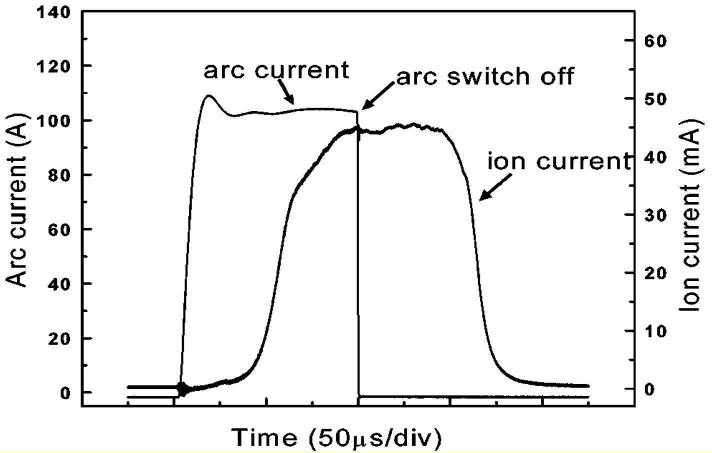
Is correct value but was criticized because he said “temperature” 500,000 K

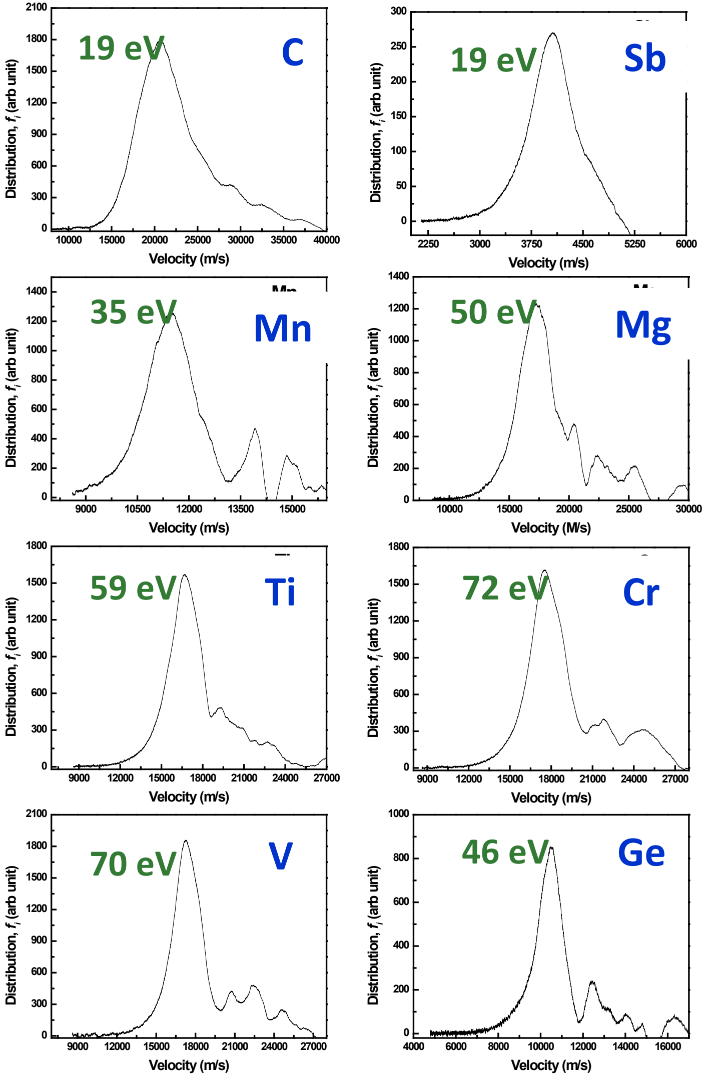
R. Tanberg, Phys. Rev., 35 (1930) 1080-1089.

Both these methods gave a vapor velocity of the order of 16x10^5 cm/sec. A temperature of around 500,000° K results when this value for the cathode vapor velocity is substituted for c in the equation: $\frac{1}{2}mc^2=3\ KT/2$.

Energy Distribution Functions for Cathodic Vacuum Arcs

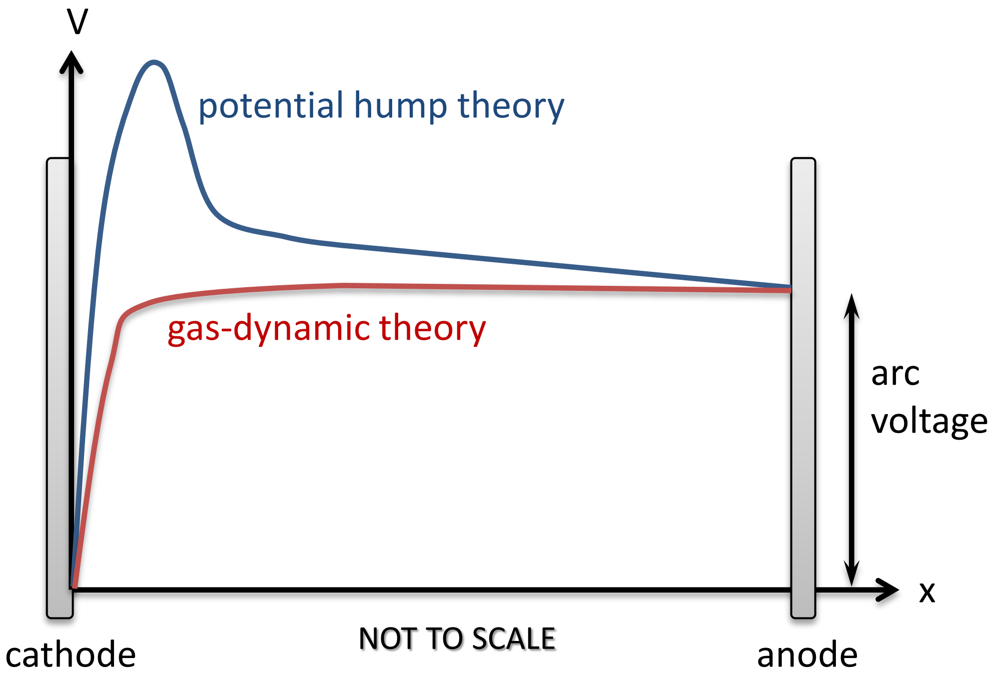
- Switch-off TOF method
- Distributions show ***one*** large peak
- → kinetic energy is almost independent of ion charge state; a pressure driven ion acceleration mechanism





A. Anders and G. Yushkov, J. Appl. Phys. **91** (2002) 4824
E. Byon and A. Anders, J. Appl. Phys. **93** (2003) 1899

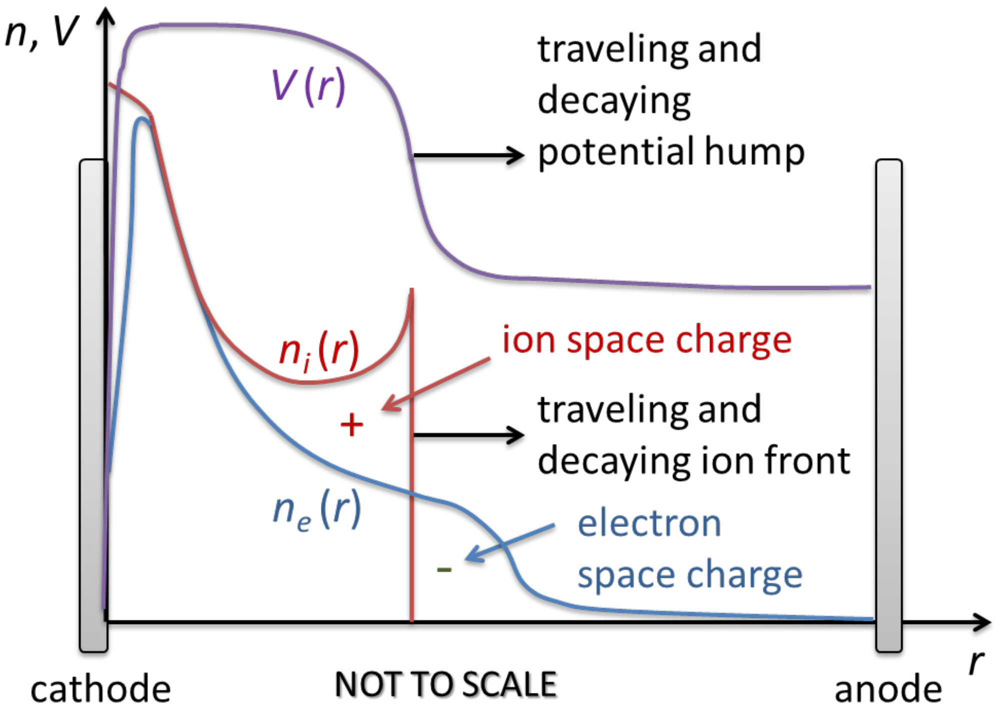
Ion acceleration near cathode spots: Potential hump vs. gas-dynamic theories



- Potential hump theory: ion acceleration in an electric field
- Gas-dynamic theory: acceleration by pressure gradients

A. Anders, Proc. 26th Int. Symp. Discharges and Electrical Insulation in Vacuum (ISDEIV 2014), Mumbai, India, 2014, pp. viii-x.

Both: Acceleration by gas-dynamic and electric field acceleration

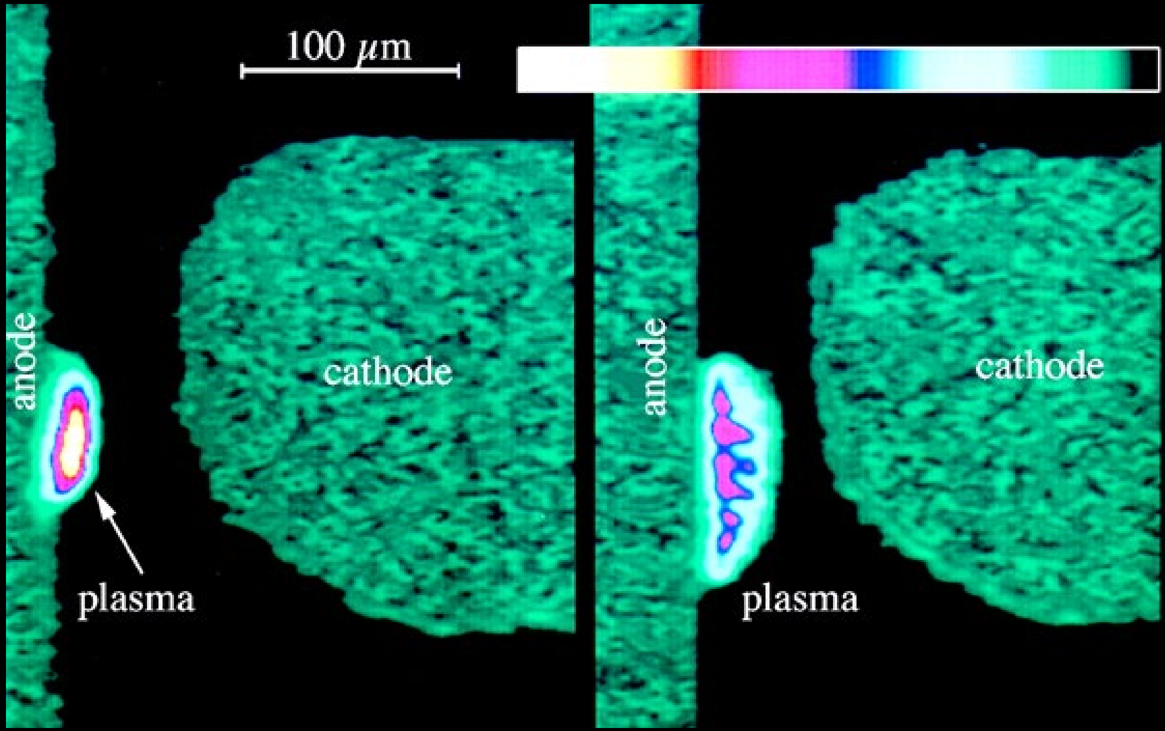


- Traveling and decaying potential humps
- Gradients are associated with the non-stationary nature of plasma formation
- Potential humps are stochastic
- Quantitative model remains to be developed

A. Anders, Proc. 26th Int. Symp. Discharges and Electrical Insulation in Vacuum (ISDEIV 2014), Mumbai, India, 2014, pp. viii-x.

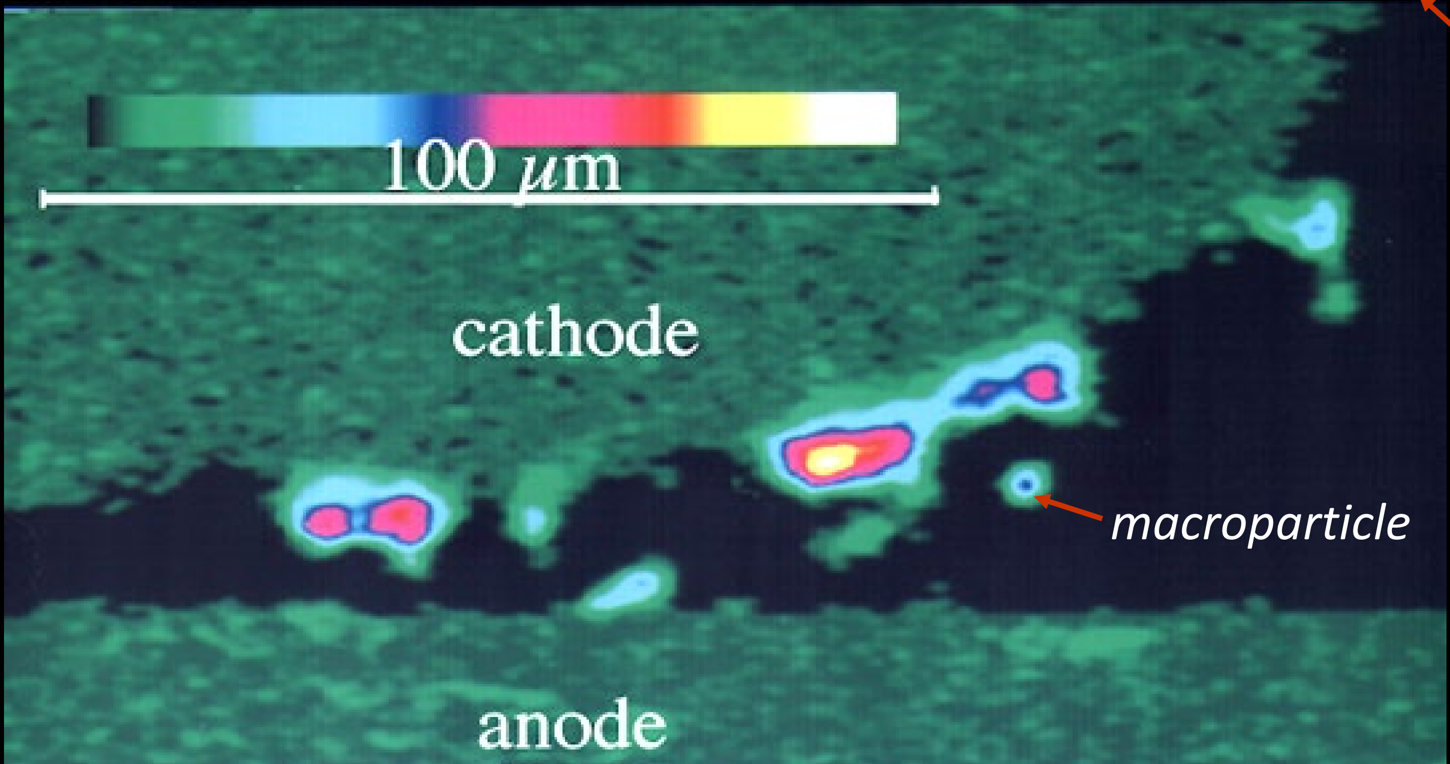
Cathodic Arc Plasmas: Observation of Gap Breakdown

- Laser absorption photography of vacuum breakdown event, Cu, 100 A, Δt between pictures 3 ns
- Spark phase of the arc: voltage is still high



A. Anders *et al*, IEEE Trans. Plasma Sci. 20 (1992) 466-472; and IEEE Trans. Plasma Sci. 24 (1996) 69.

Cathodic Arc Plasmas: Spot & "Spot Fragment" Formation



Absorption photography, copper cathode, UHV conditions, arc current 90 A, 232 ns after ignition, exposure time 0.4 ns

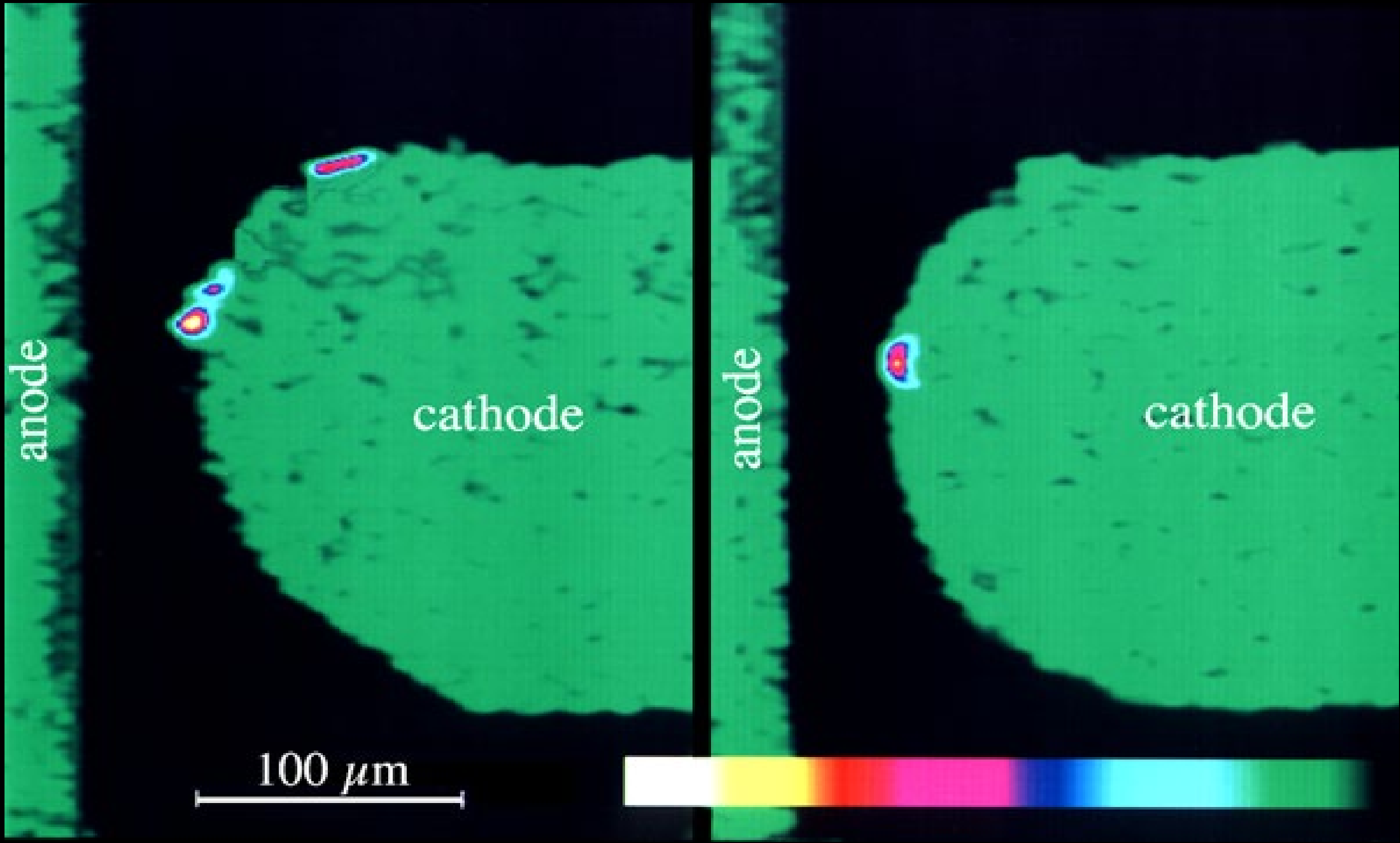
Terminology "fragment" not needed if one thinks of a fractal.

Example: visually one spot but containing "fragments"

A. Anders *et al*, IEEE Trans. Plasma Sci. 20 (1992) 466-472; and IEEE Trans. Plasma Sci. 24 (1996) 69.

Cathode Spot Dynamics

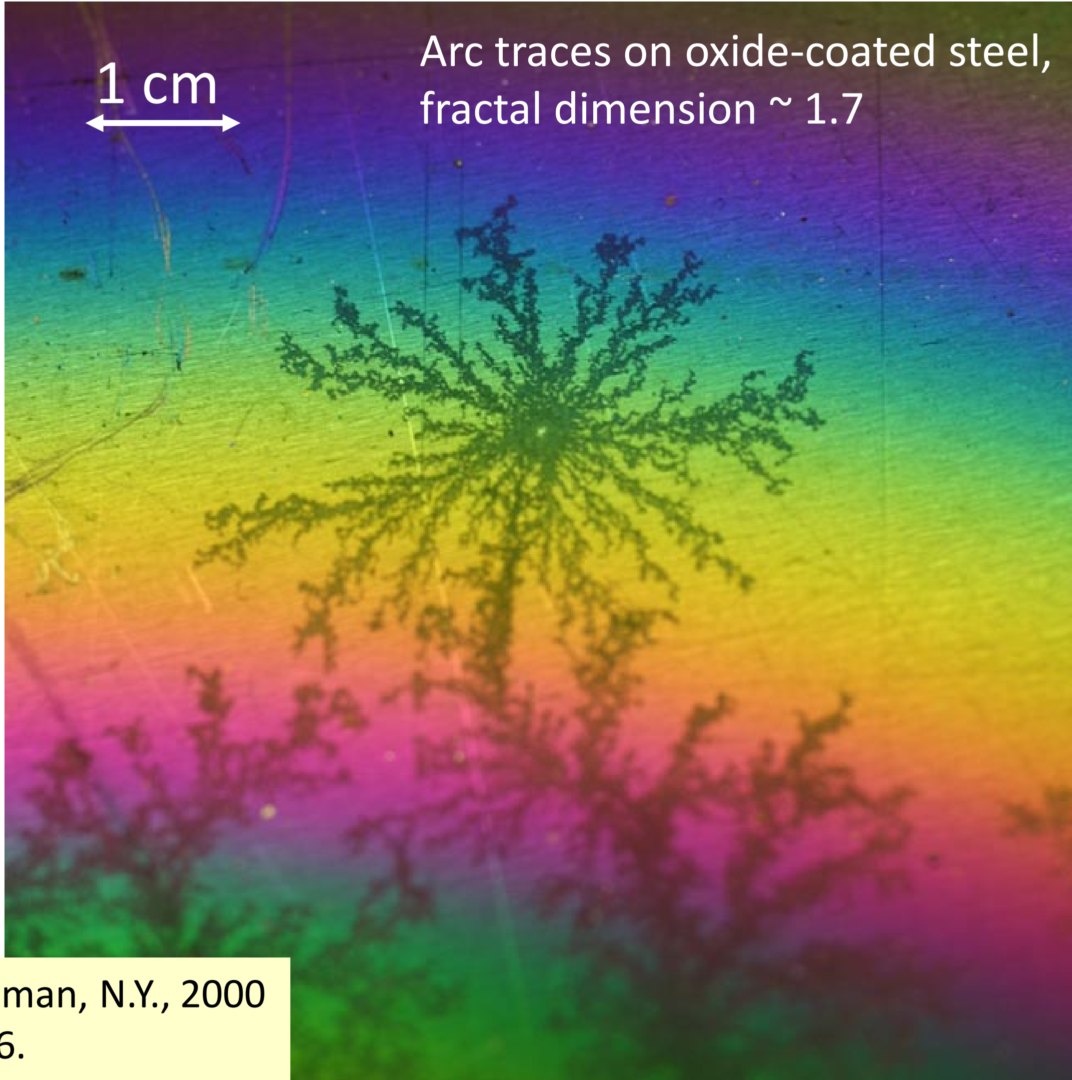
development of cathode spots, observed by absorption photography, Cu, 100 A, Δt between pictures 3ns



A. Anders *et al*, IEEE Trans. Plasma Sci. 20 (1992) 466-472; and IEEE Trans. Plasma Sci. 24 (1996) 69.

The Fractal Nature of Cathode Spots

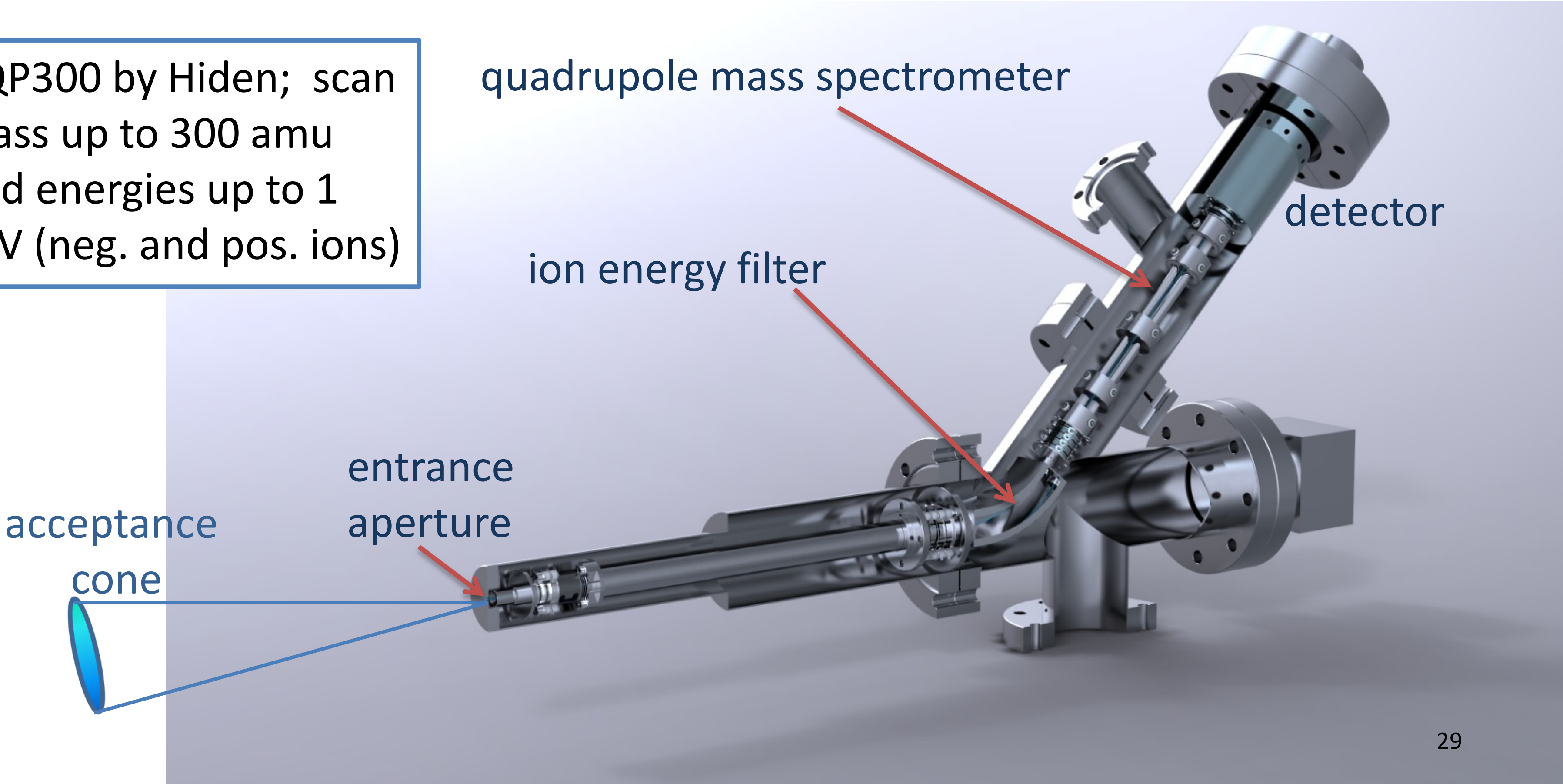
- An object ("fractal") is self-similar (= invariant with scaling) if it is reproduced by magnifying some portion of it.
- Self-similarity may be discrete or continuous, deterministic or probabilistic.
- Self-similarity can be mathematically exact or only approximate and asymptotical.
- Physical fractals have lower and upper scaling limits



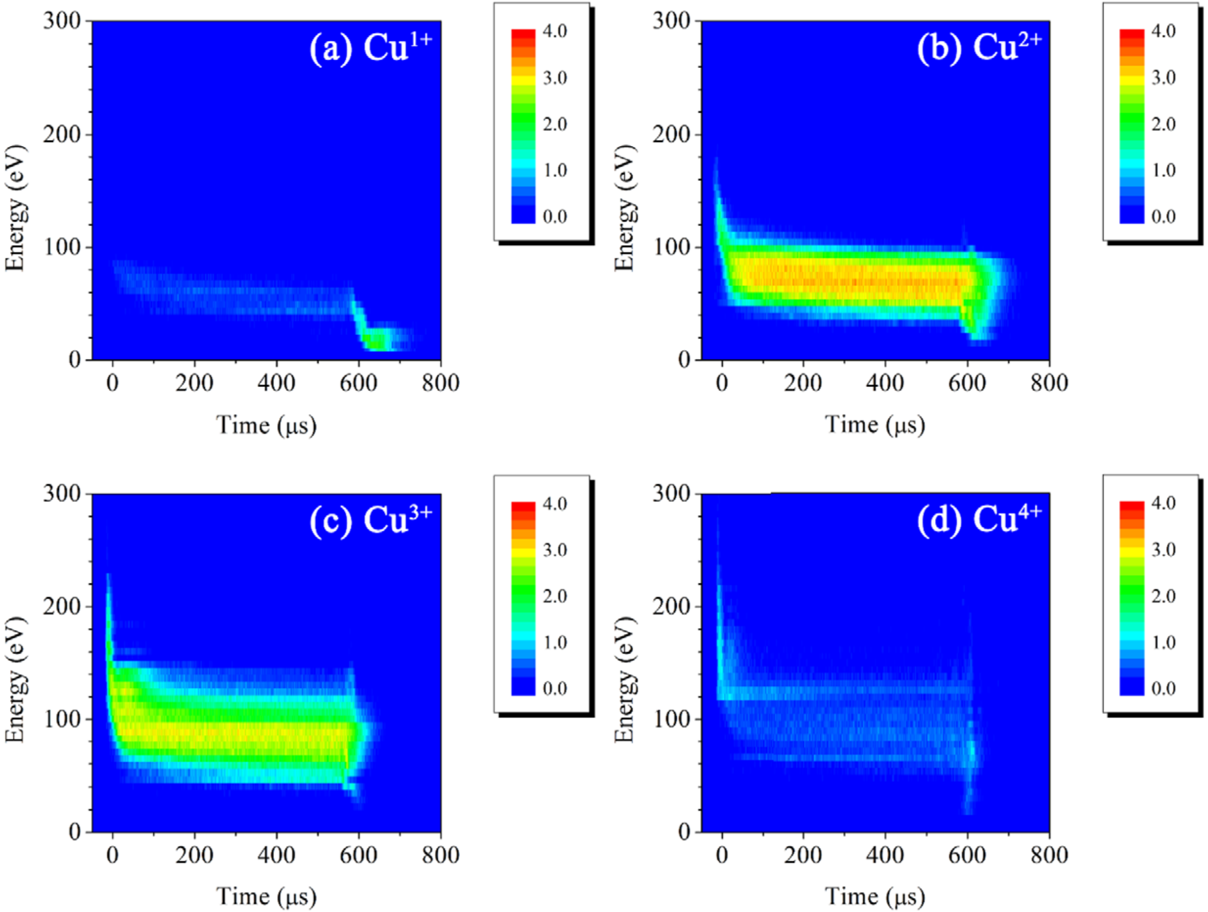
M. Schroeder, Fractals, Chaos, Power Laws, Freeman, N.Y., 2000
A. Anders, IEEE Trans. Plasma Sci. **33** (2005) 1456.

Ion flux spectrometry via ion energy/charge filter plus quadrupole mass/charge filter

EQP300 by Hiden; scan mass up to 300 amu and energies up to 1 keV (neg. and pos. ions)



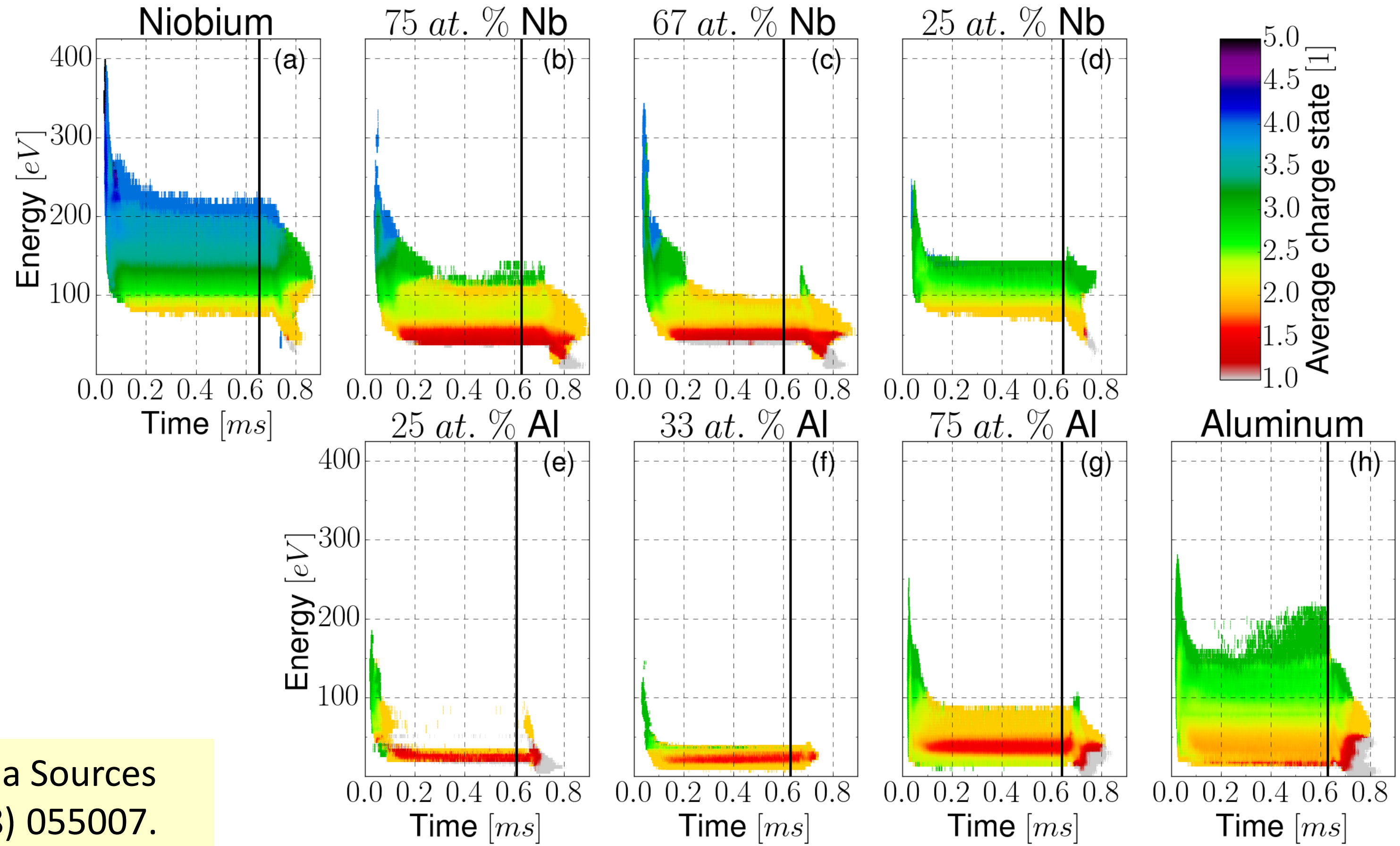
Charge-state and time-resolved energy distribution functions for cathodic vacuum arcs



- multiply charged ions
- higher at beginning of discharge
- steady-state afer ~ 100 μs
- fluctuating data, results show average over many measurements

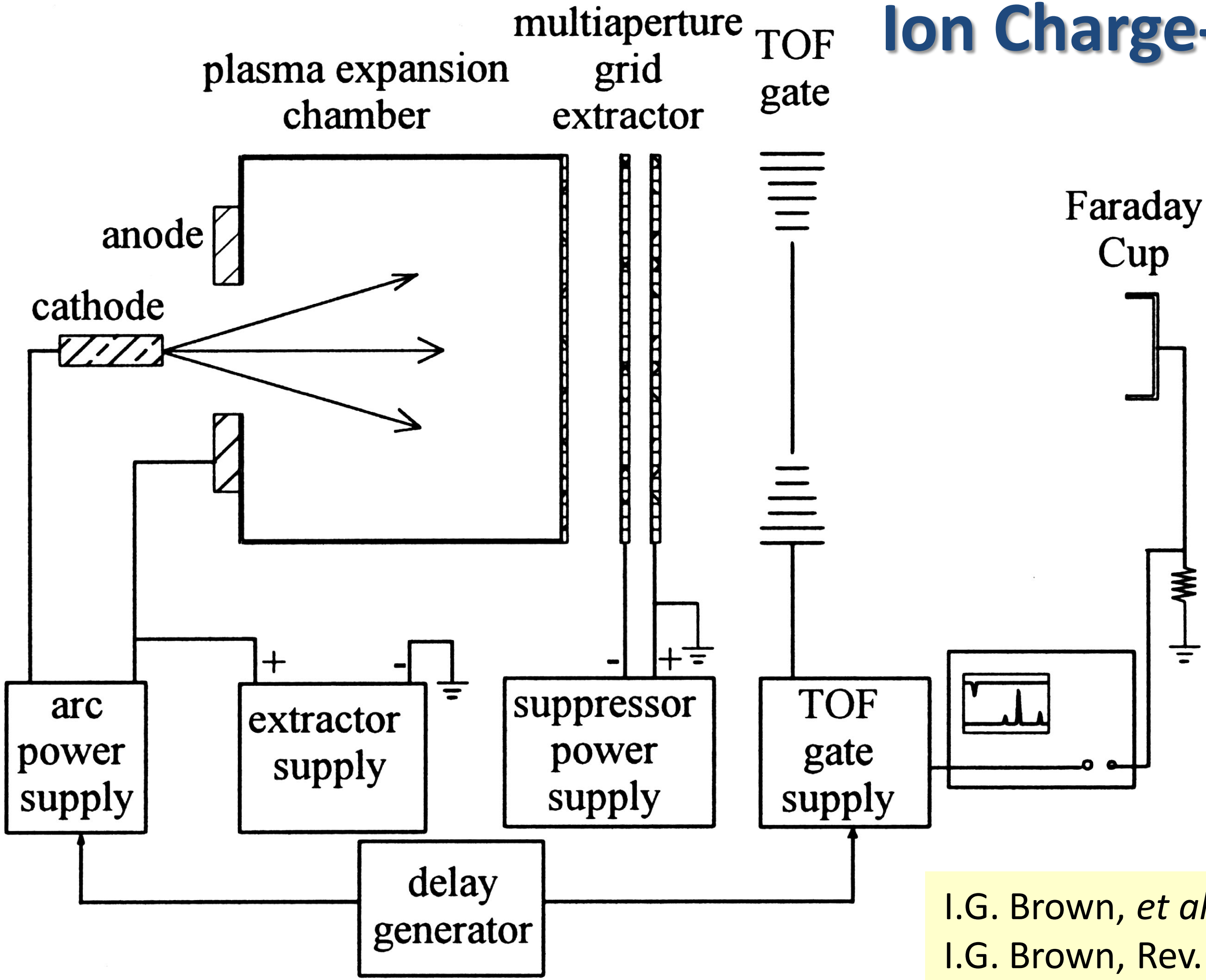
K. Tanaka, *et al.*, Plasma Sources Sci. Technol. **24** (2015) 045010.

Charge-state and time-resolved energy distribution functions for cathodic vacuum arcs: Nb-Al cathodes

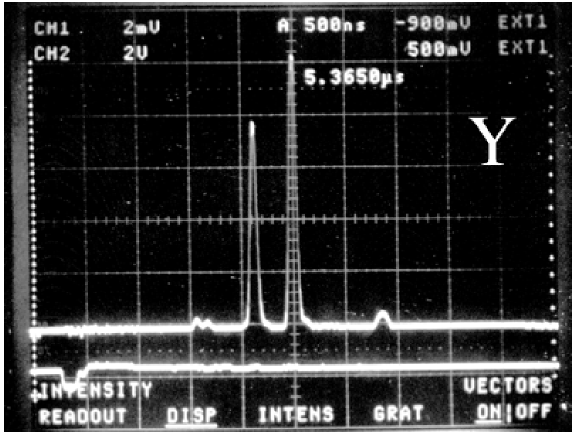


S. Zöhrer, *et al.* Plasma Sources Sci. Technol. **27** (2018) 055007.

Ion Charge-State Spectrometry via TOF



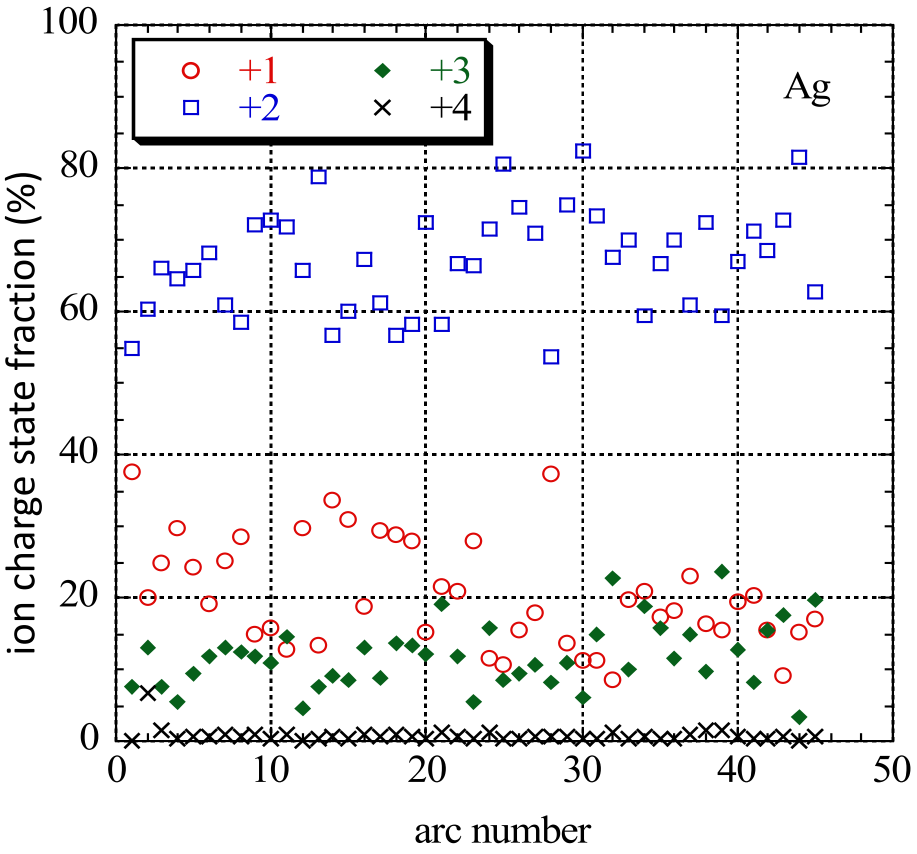
time-of-flight (TOF) method



I.G. Brown, *et al.*, Rev. Sci. Instrum. 58 (1987) 1589.
I.G. Brown, Rev. Sci. Instrum., 65 (1994) 3061.

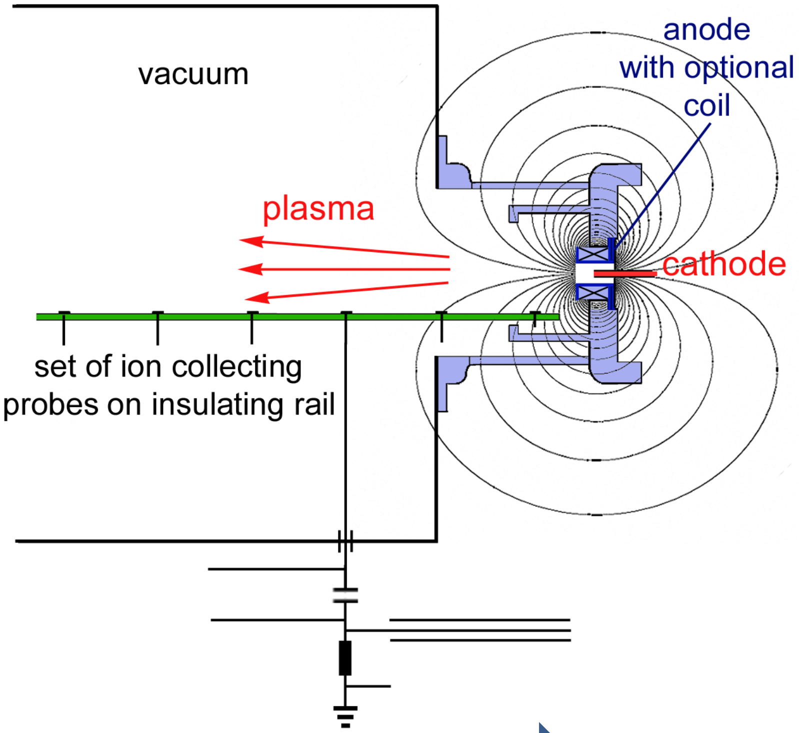
Fluctuations or “noise” can be observed in all parameters

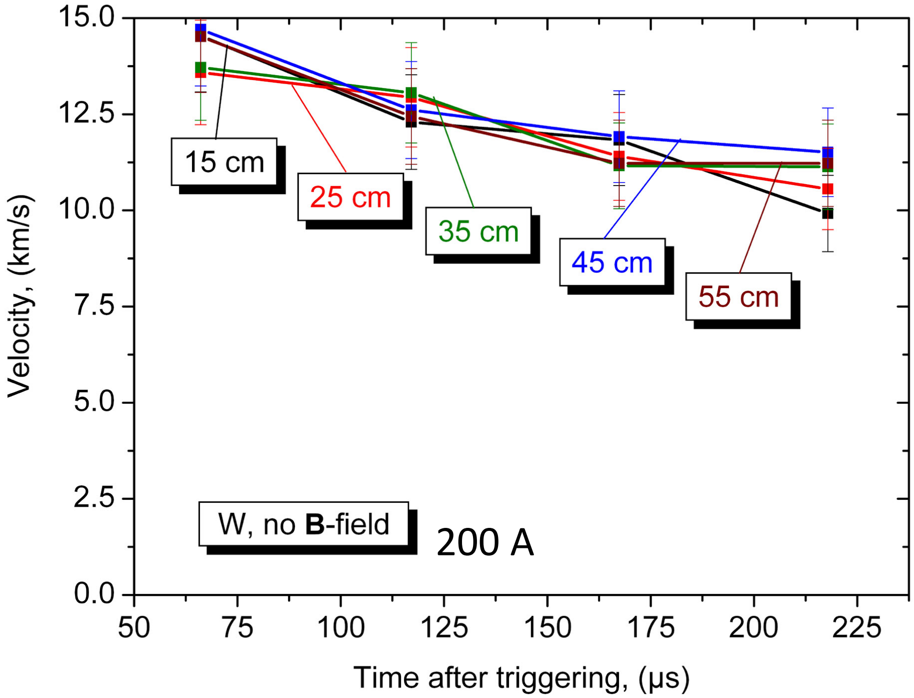
- ❑ Fluctuations (or “noise”) is due to non-stationary plasma formation
- ❑ “Noise” is of little or no concern for plasma deposition (averaging effect) but essential for operation of the arc
- ❑ Example here: Ion charge state sampling of a pulsed silver arc



A. Anders, IEEE Trans. Plasma Sci. **27** (1999) 1060.

Evolution of ion velocities (time, distance from cathode)

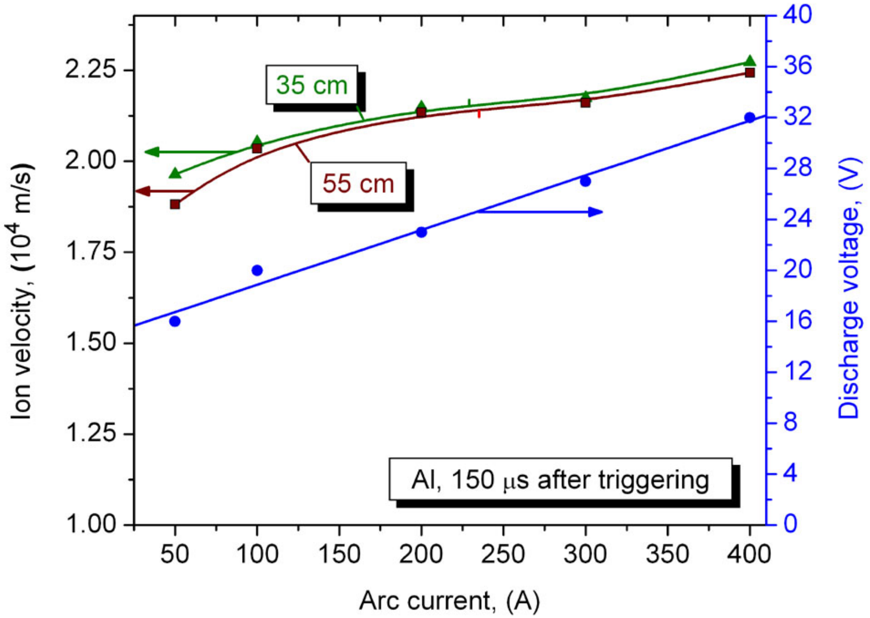


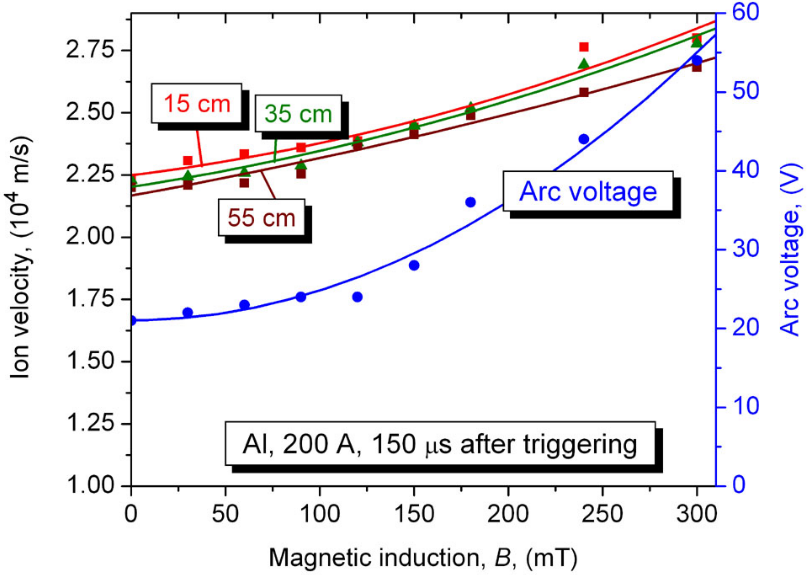


- average velocity reduces slightly as time elapsed after arc start
- velocity is essentially constant as plasma propagates away from the cathode

S. Hohenbild, et al., J. Phys. D: Appl. Phys. **41** (2008) 205210.

Evolution of ion velocities (arc current, magnetic field)

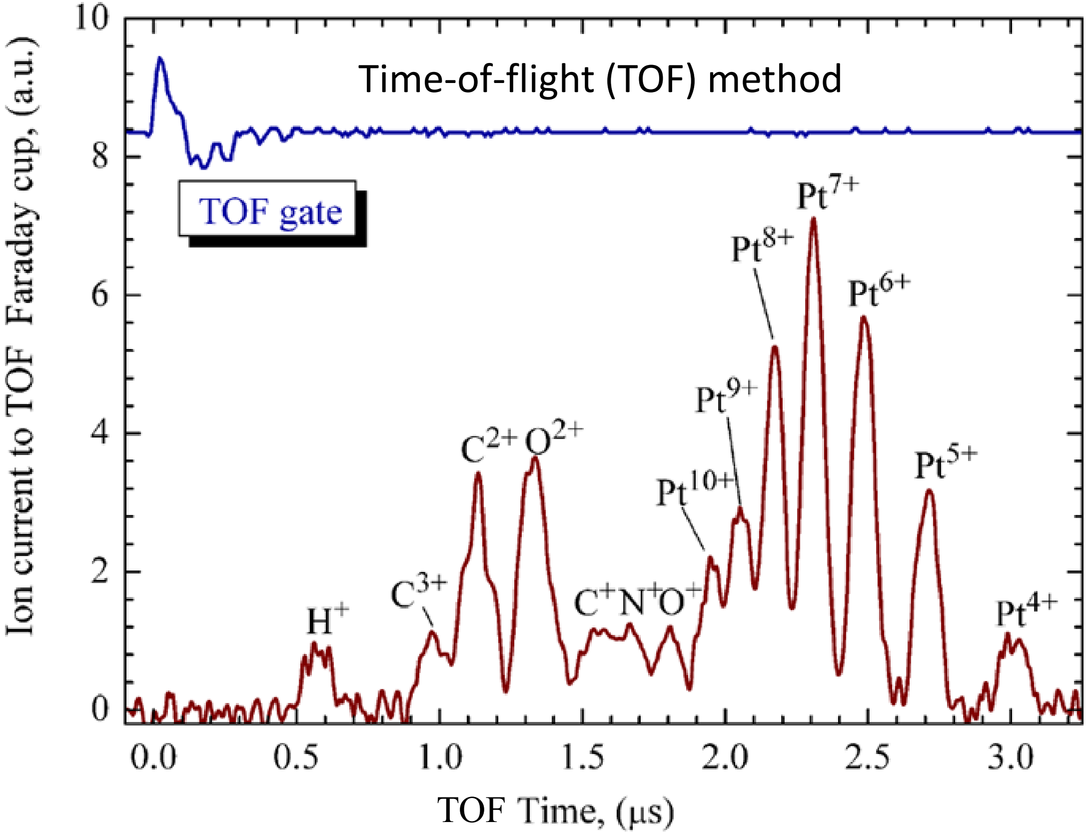




- ion velocity slight increases with arc current → most of the acceleration happened at cathode spot, supplemented by acceleration in the near-cathode density gradients →
- portion of additional near-cathode-acceleration can be enhanced by magnetic field (power enhancement)

S. Hohenbild, et al., J. Phys. D: Appl. Phys. **41** (2008) 205210.

Enhancing Ion Charge States



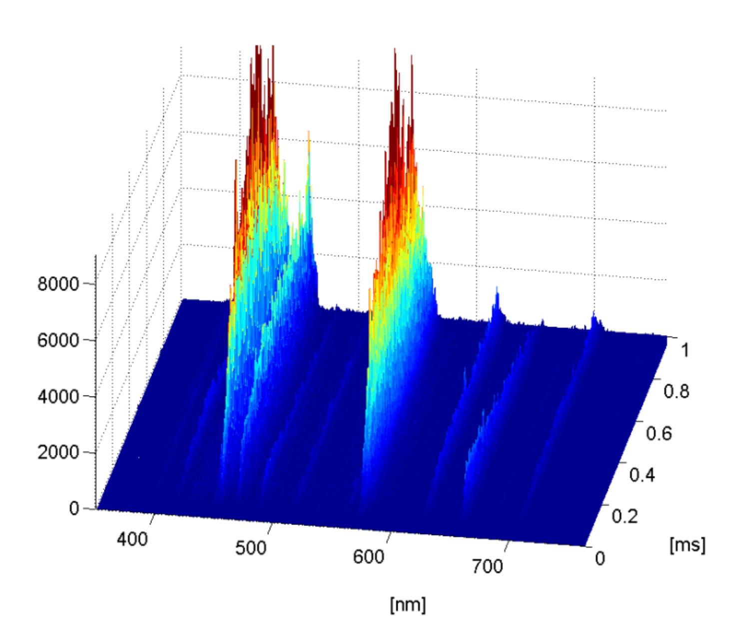
- 2+ and 3+ is typical for most metals
- using short pulses and a magnetic field → up to 10^+ has been detected, indicative for high electron temperature and many collisions in dense plasma

G. Yushkov and A. Anders,
Appl. Phys. Lett. **92** (2008) 041502

Reducing Ion Charge States

Sticking coefficient
is less than unity

optical spectrum: Al <u>neutral</u> lines appear and increase with time



neutrals lead to charge exchange

$$M^{Q+} + M^{0} \rightarrow M^{(Q-1)+} + M^{+}$$

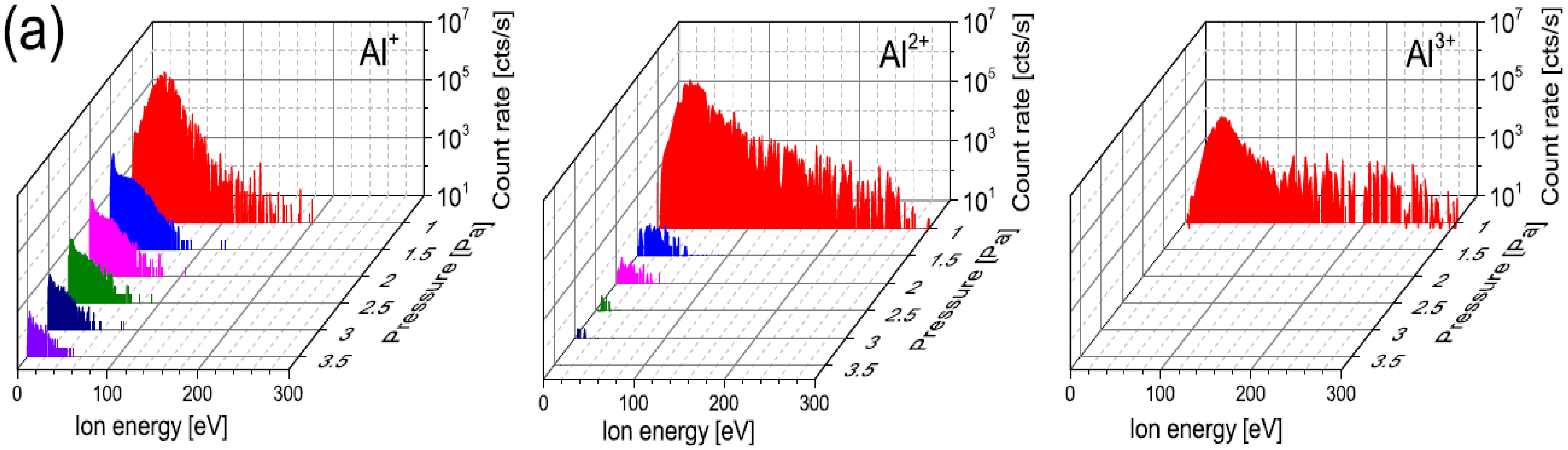
and thus to a reduction of higher charge states

P.A. Ni, A. Anders, J. Phys. D: Appl. Phys. **43** (2010) 135201.

Introduction of reactive gas reduces ion charge states and ion energies

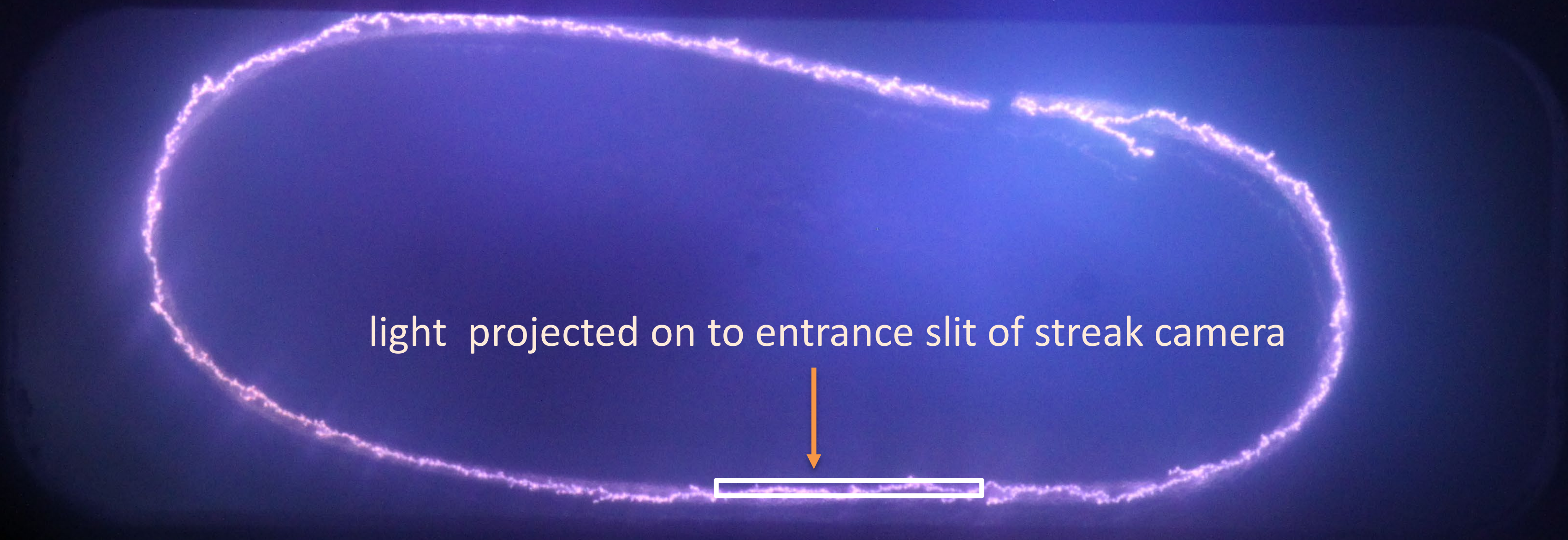
Example here: Al cathode in O_2 atmosphere, effects due to

- collisions, mostly charge exchange collisions, and
- changes of the chemical composition of the cathode itself ("poisoning")



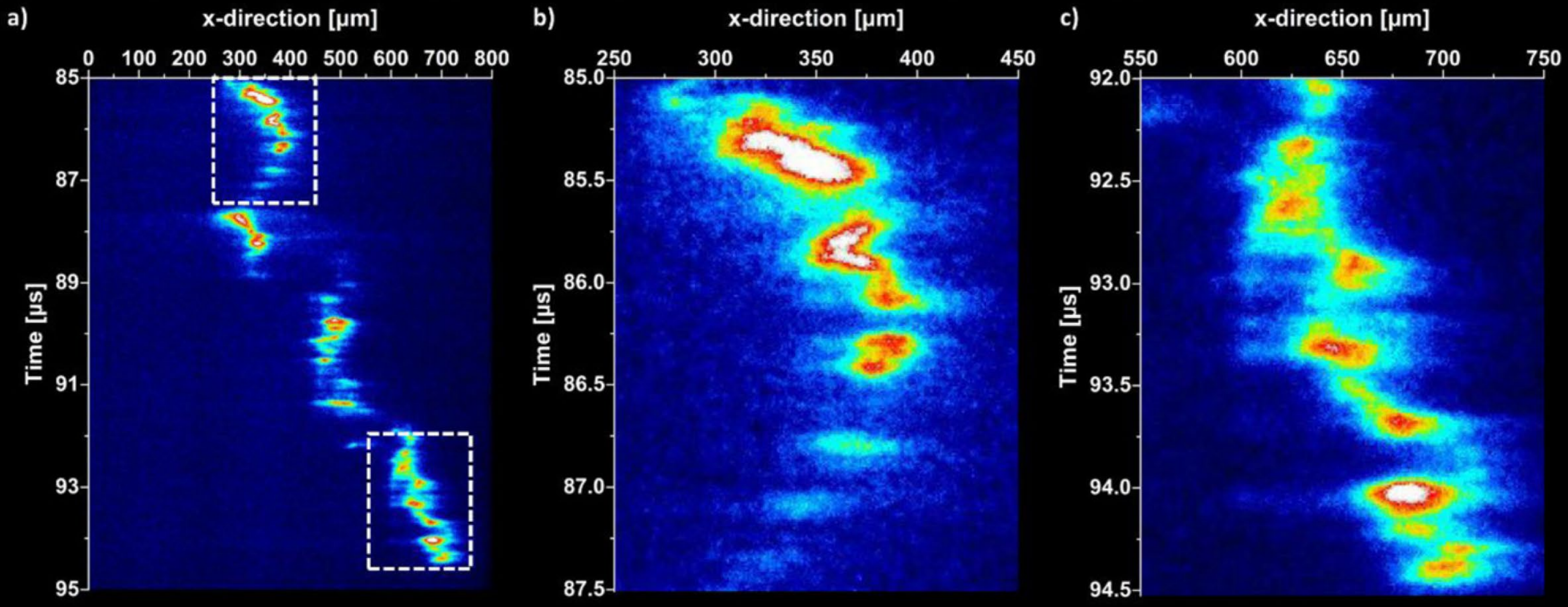
R. Franz, *et al.*, Surf. Coat. Technol. **272** (2015) 309.

Further studies of fluctuations using a “magnetically steered” arc: Search for “characteristic times



(slit not to scale)

Study of Fluctations: Cathode Spots are Fractals

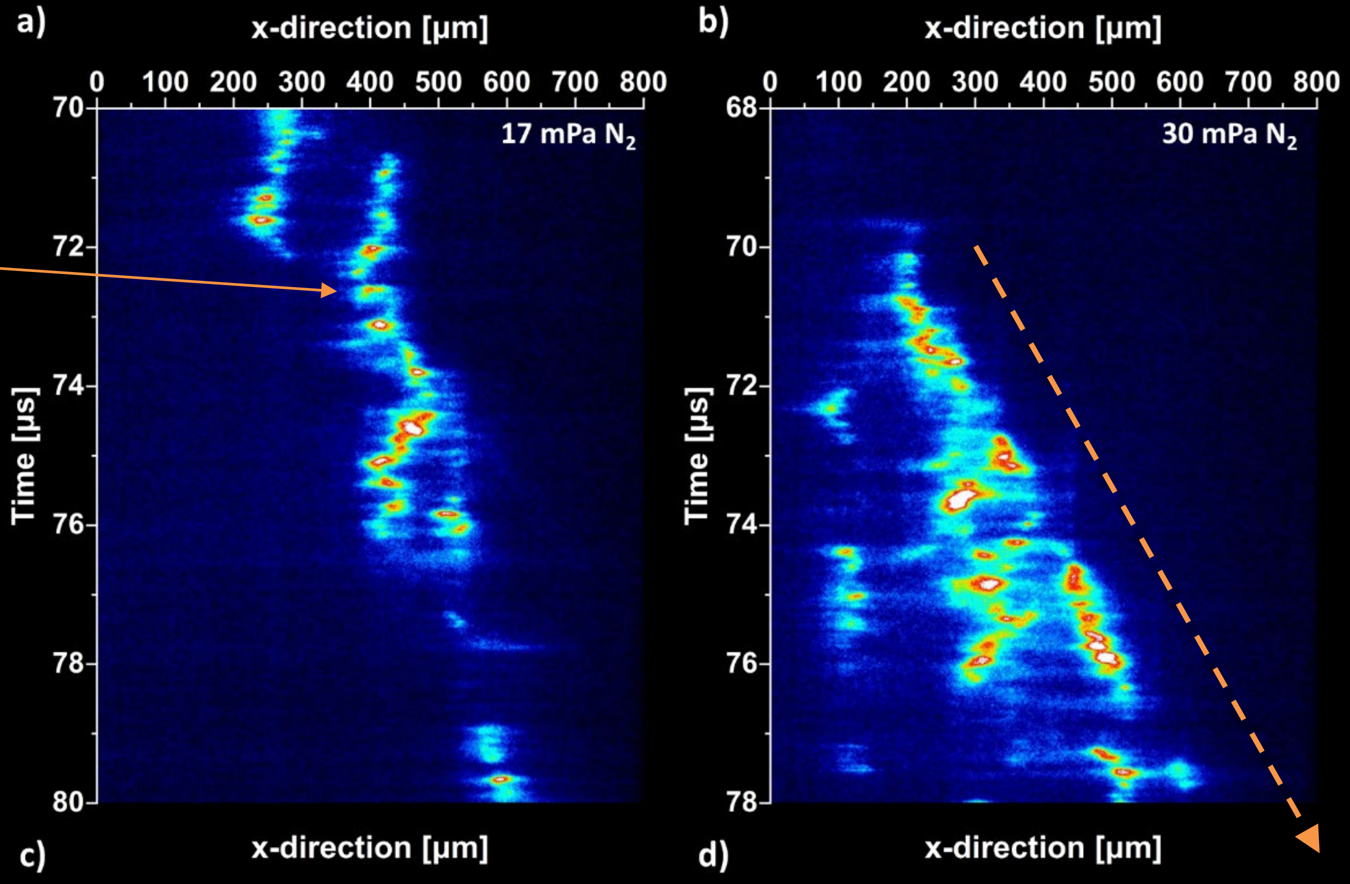


FFT analysis→ spots do not show periodic but self-similar (fractal) features

K. Oh, et al., Plasma Sources Sci. Technol., 30 (2021) 095005.

Study of Fluctations: Cathode Spots as used in reactive arc-PVD

repeated ignition at about the same site

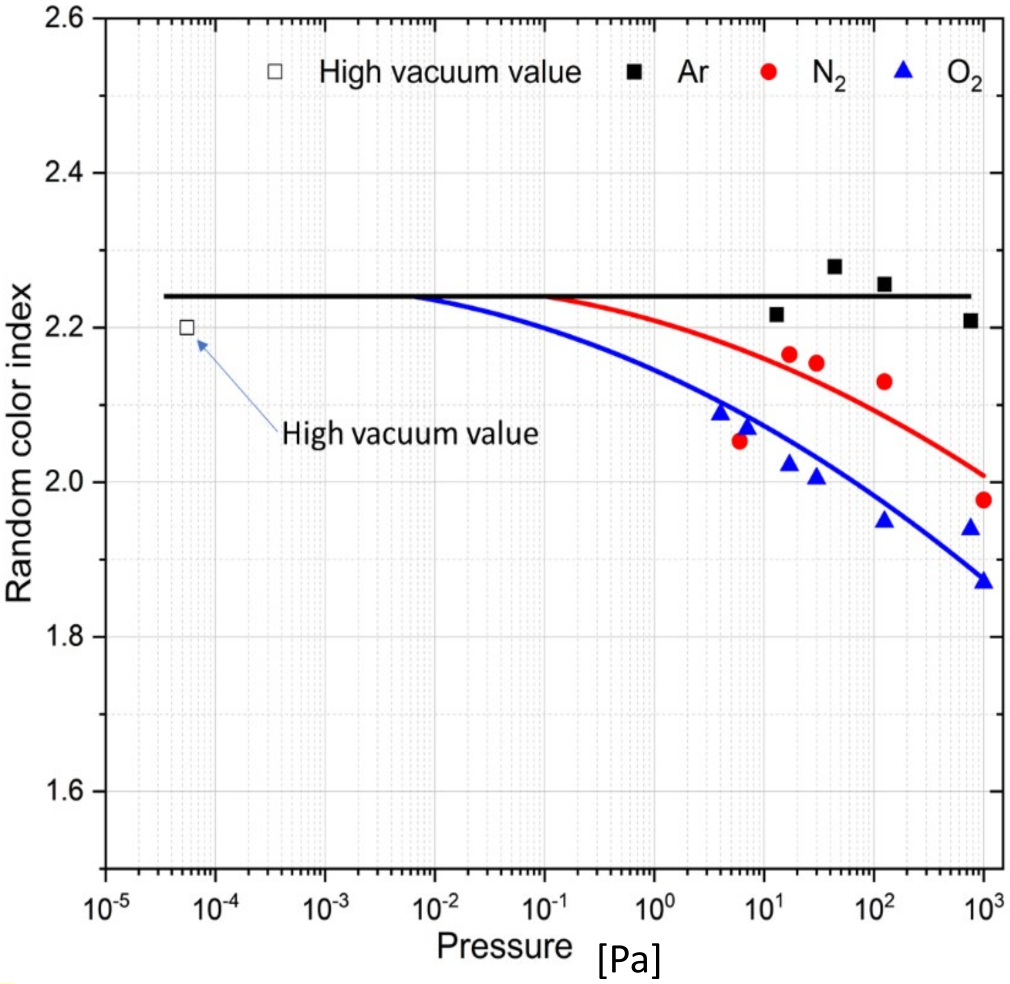


apparent retrograde motion of spots

K. Oh, *et al.*, J. Appl. Phys., **130** (2021) 183304.

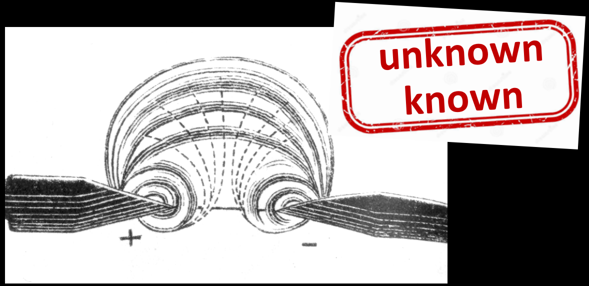
Cathodic arc spots in the presence of a reactive gas

1. fluctuations or “noise” can be quantified by Random Color Noise (RCN) index
2. FFT shows no peaks → no characteristic times of “arc cycles”
3. compound layers promote the ignition of new spots and reduce the feedback dependence on previous spots



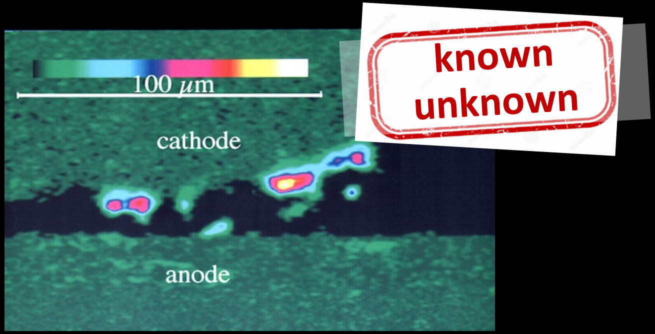
K. Oh, *et al.*, J. Appl. Phys., **130** (2021) 183304.

Summary

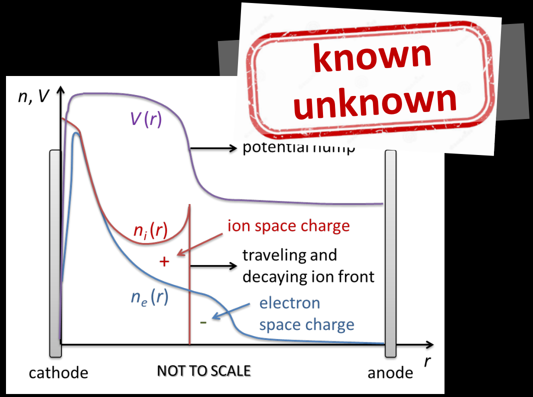


arcs and arc history

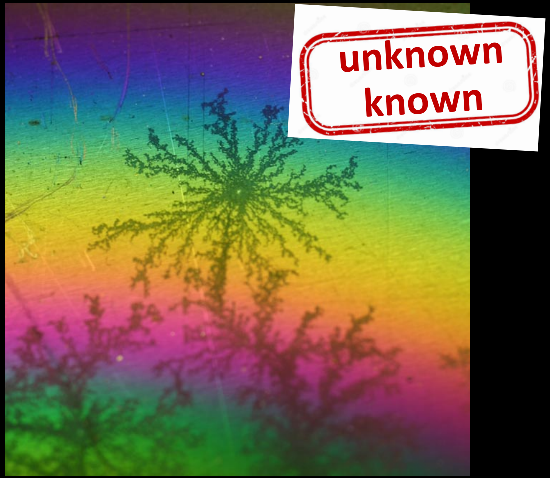
cathode spots



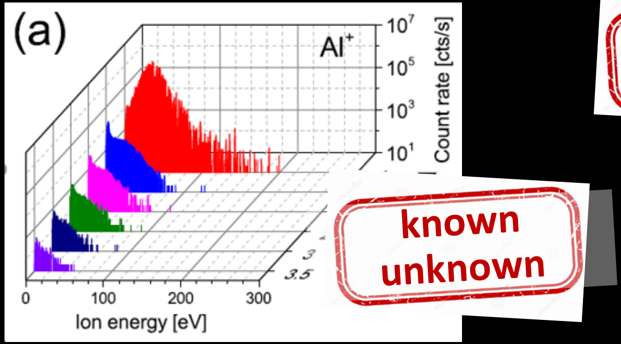
spot and “fragments”



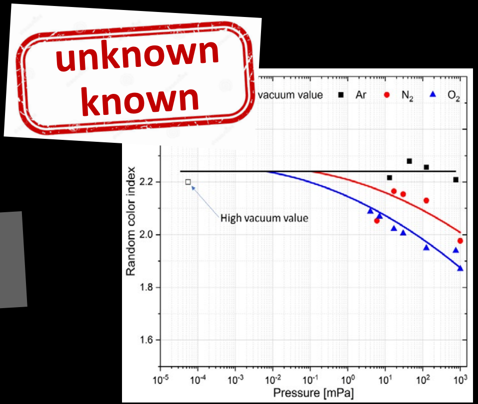
qualitative ion acceleration model



fractal nature of cathodic arcs



ion charge state and energy distribution functions



Quantification of fluctuations