SPECTROSCOPIC METHOD FOR NITROGEN IMPURITY ESTIMATION IN HELIUM ATMOSPHERIC DISCHARGE

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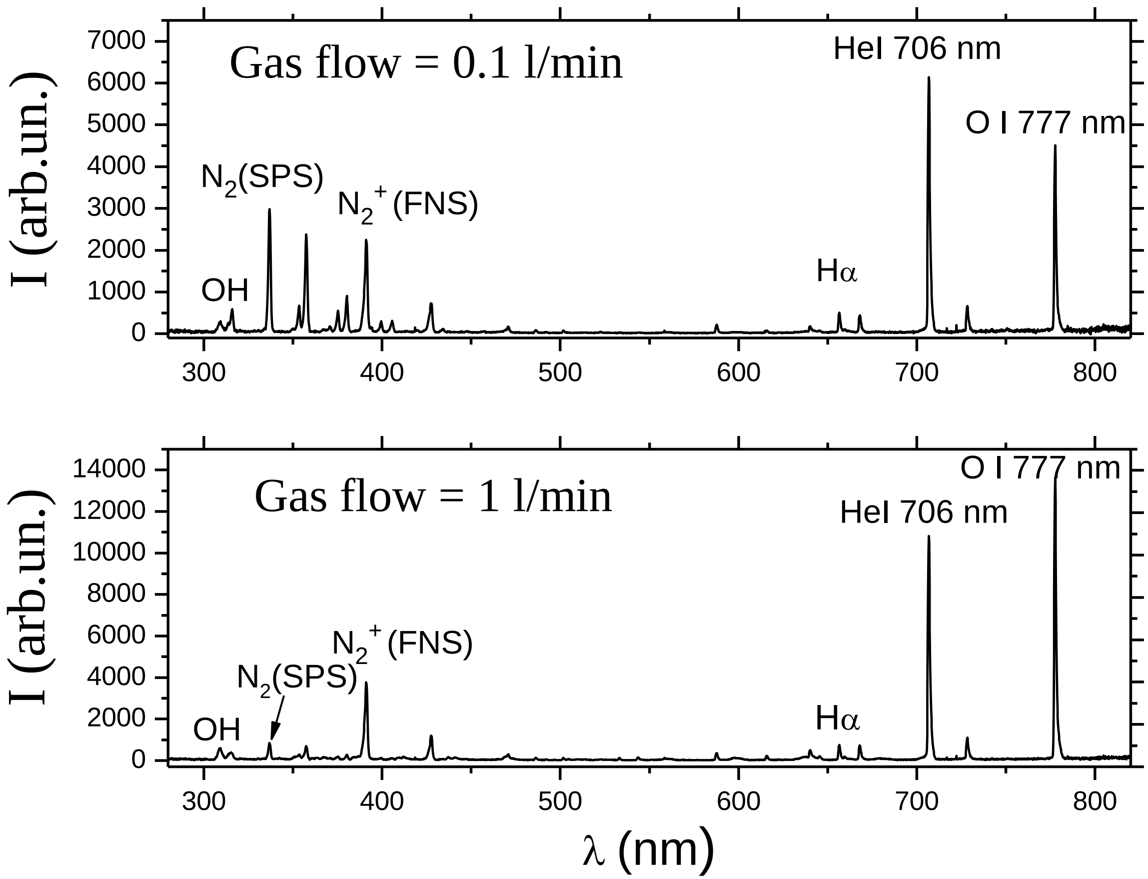
Cold Atmospheric pressure discharges

- Non-thermal or cold discharges, are presently most investigated and most promising laboratory plasma sources.
- In the last two decades they have been extensively studied both theoretically and experimentally.
- Plasma is strongly out of equilibrium with electron temperature of the order 10000 K while ions and atoms are at close to room temperature (therefore *cold*).
- **Dielectric barrier discharges (DBDs)** operating in noble gases mostly He and Ar.

Why are impurities important in atmospheric discharges?

- Gas impurities within the working gas are crucial for barrier discharge operation mostly due to metastable processes.
- Numerous models have shown the influence of gas impurity level on discharge parameters.
- Impurities originate mostly from the air protruding through the chamber, gas supply system, but also some traces are always in the cylinder
- The impurity composition is mostly N_2 but also O_2 and H_2O.
- There is a necessity for a spectroscopic measurement of impurity level

- Our method is based on the intensity ratio of prominent nitrogen molecular band and strong helium line.
- N_2 ($C^3\Pi u$–$B^3\Pi g$, 0-0) at 337 nm, and the
- He I (3^3S-2^3P) at 706 nm .



Collisional-radiative model:

- Collisional-radiative model was developed, and a functional dependance of intensity ratio on impurity at a given reduced electric field was numerically obtained.
- The ratio is obtained from the number density of exited species:

$$R_{337/706} = \frac{I_{N_2\_337nm}}{I_{He\_706\,nm}} = \frac{h\nu_{337} \cdot A_{337} \cdot [N_2\,(C)]}{h\nu_{706} \cdot A_{706} \cdot [He3^3S]} = f\left(\frac{[N_2]}{[He]}\right) = f\left(N_2[ppm]\right)$$

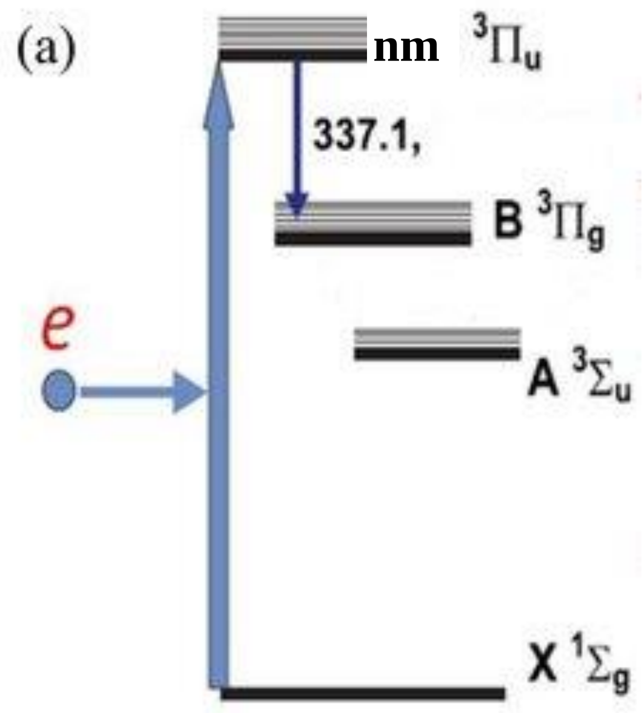
N2(C)

- In helium plasma, the nitrogen excited $N_2(C)$ density can be calculated from processes involving electron excitation, N_4^+ recombination, and pooling from $N_2(A)$ metastable.
- When nitrogen is present as an impurity, the metastable pulling is negligible.

- $e + N_4^+ \rightarrow N_2(C) + N_2$
- $e + N_2 \rightarrow N_2(C)$

- Steady state equation is valid in our conditions:

$$N_2(C) = \frac{k_1 n_e [N_2] + k_2 n_e [N_4^+]}{k_3 + k_4 [N_2] + k_5 [He]}$$

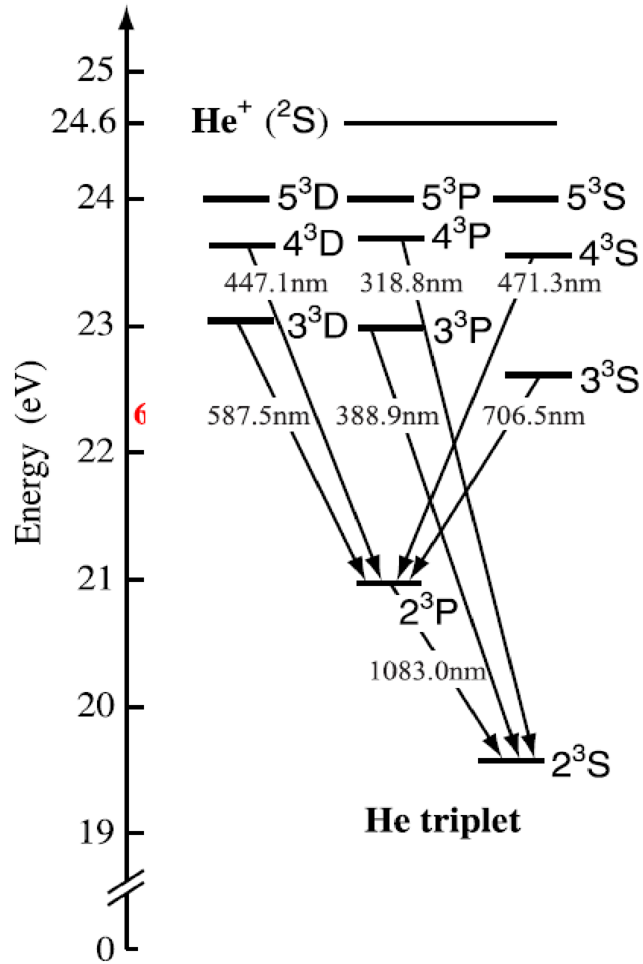


He 3^3S

- System of three time-dependent equations for the interconnected levels 3^3D, 3^3P and 3^3S are:

$$\frac{d[\mathrm{He}^*]}{dt} = k_e n_e[\mathrm{He}] + k_{em} n_e [\mathrm{He_m}] + k_{ext}[\mathrm{He}^{**}][\mathrm{He}] - [\mathrm{He}^*]\cdot\left(A + k_q[\mathrm{He}]\right)$$

- Electron excitation from the ground level and from the metastable He 2^3S
- excitation transfer from He 3^3D and He 3^3P
- The electron excitation rate constant from the ground level He (k_e) and metastable Hem (k_{em}) are obtained from the BOLSIG– solver.



- Electron collision excitation rates are obtained from Boltzman solver and depend on E/N.

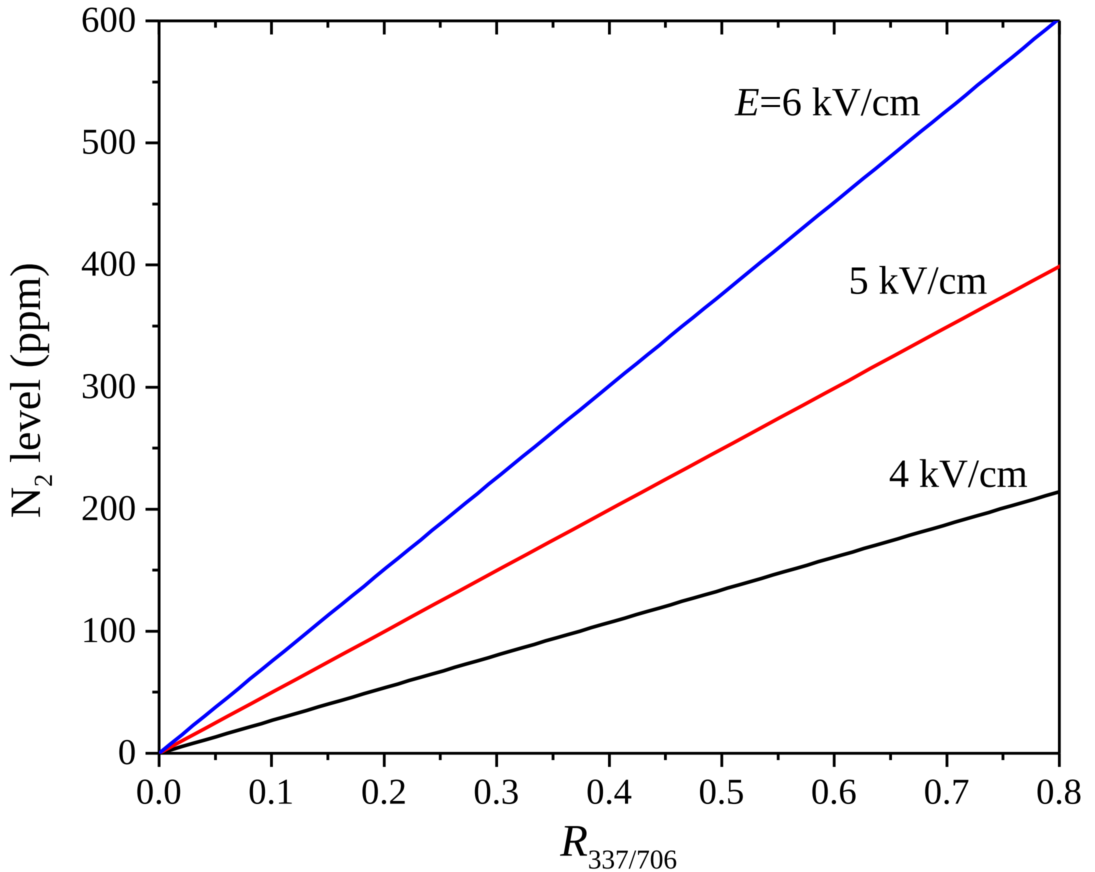
Process		Value
	deexcitation coefficients	
Spontaneous emission He:	$3^3S \rightarrow 2^3P$	$A_{706} = 2.75\times10^7\ s^{-1}$
	$3^3P \rightarrow 2^3S$	$9.47\times10^6\ s^{-1}$
	$3^3D \rightarrow 2^3S$	$6.56\times10^7\ s^{-1}$
N_2:	$N_2(C) \rightarrow N_2(B)$	$k_3 = 2.75\times10^7\ s^{-1}$
	SPS (0-0)	$A_{337} = 1.31\times10^7\ s^{-1}$
	Rate constants	
Excitation transfer:	$3^1P \rightarrow 3^1S$	$k_{ext} = 4.89\times10^{-10}\ cm^3\ s^{-1}$
	$3^1D \rightarrow 3^1P$	$k_{ext} = 1.81\times10^{-10}\ cm^3\ s^{-1}$
Collisional quenching:	$He(3^3S) + He$	$k_q=0.53\times10^{-11}\ cm^3\ s^{-1}$
	$He(3^3P) + He$	$9.8\times10^{-11}\ cm^3\ s^{-1}$
	$He(3^3D) + He$	$2.4\times10^{-11}\ cm^3\ s^{-1}$
	$N_2(C) + N_2$	$k_4 = 1.14\times10^{-11}\ cm^3\ s^{-1}$
	$N_2(C) + He$	$k_5 = 1\times10^{-12}\ cm^3\ s^{-1}$
Electron recombinaton:	$e + N_4^+ \rightarrow N_2(C)$	$k_2 = 4.6\times10^{-6}\ cm^3\ s^{-1}$

Resulting equation:

$$N_2[ppm] = \left(-327 + 9.6 \times \left(\frac{E}{N} \right)^{1.47} \right) \times R_{337/706}$$

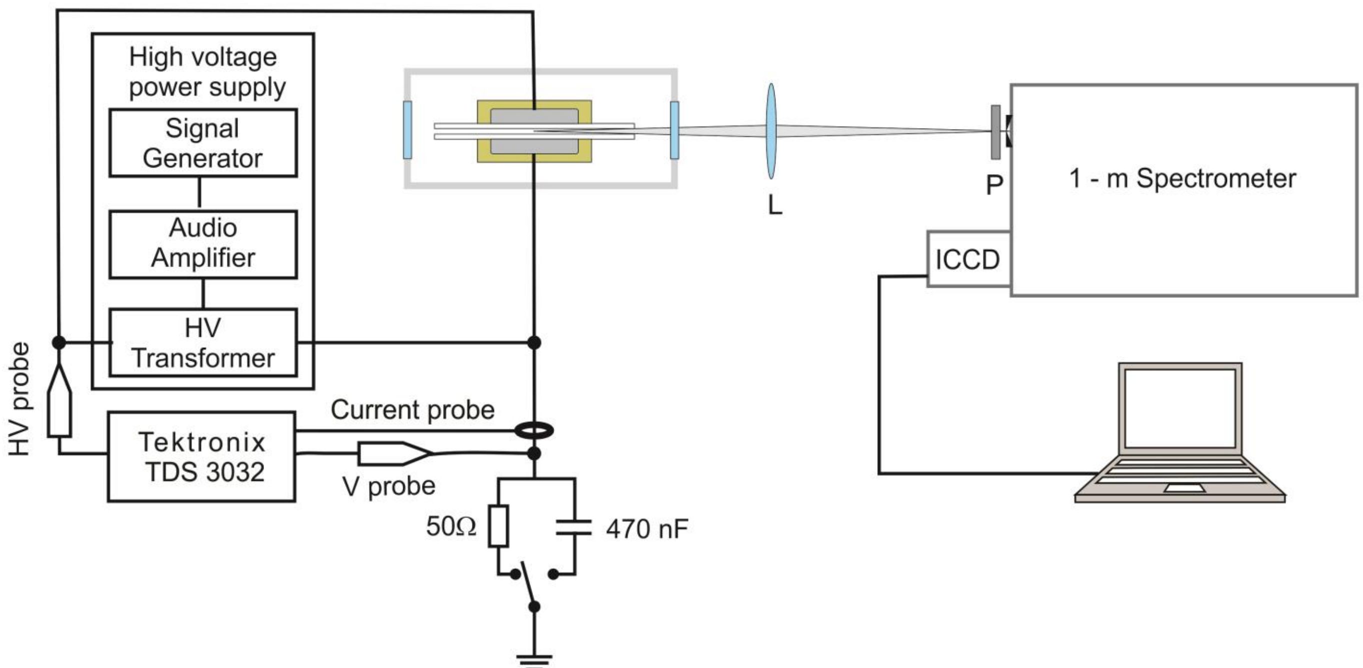
Because of metastable excitaion it is is valid only E/N > 8 Td

Linear, but field sensitive!



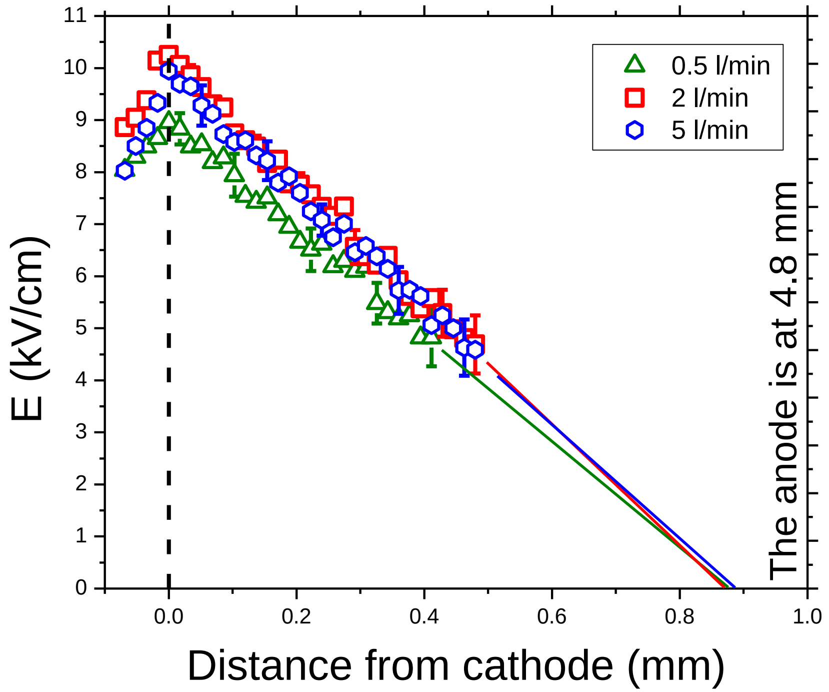
Experiment: DBD with varying helium flow

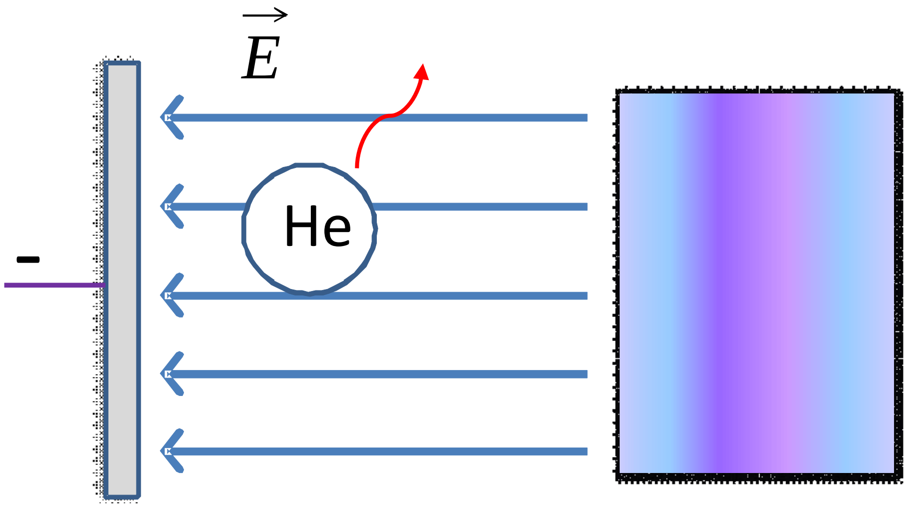
- Investigation of discharge behavior with change of helium gas flow
- Electrical and spectroscopic measurements where performed
- Strong change of electric properties and line intensities can be attributed to different causes
- One possible explanation was the decrease of impurities with stronger gas flow

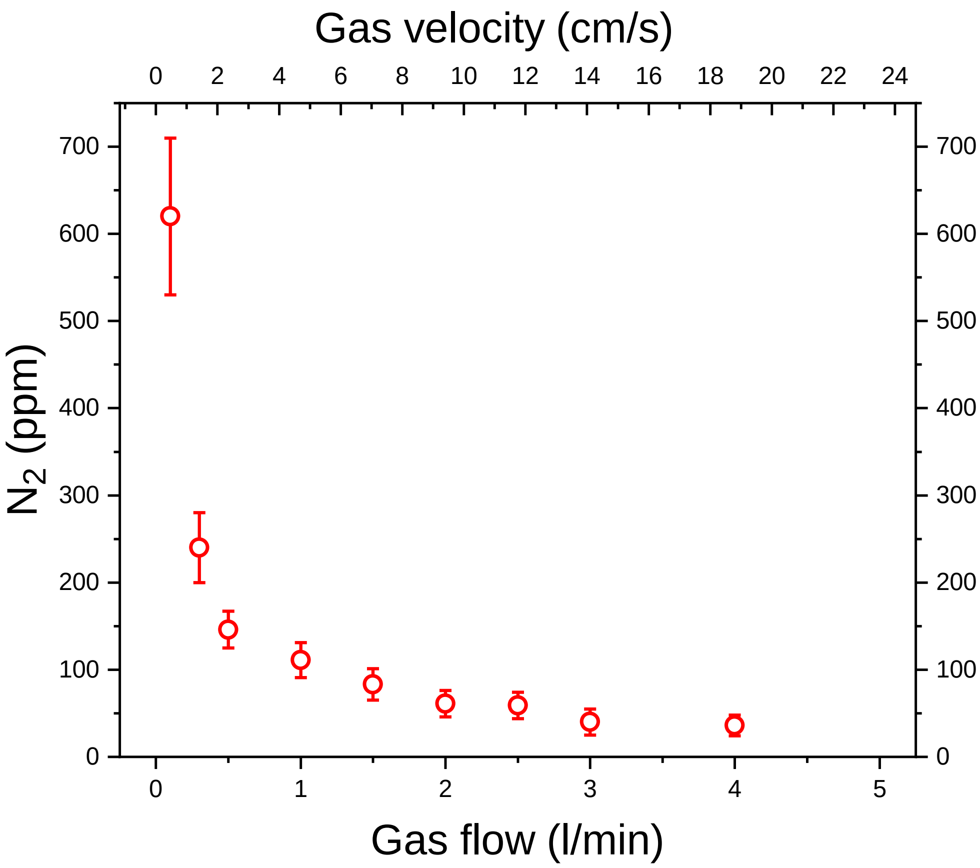


Experiment

- Electric field was measured using Stark spectroscopy







- Uncertainty is high at high N2 levels
- Minimum value corresponds to the level from the gas cylinder
- The obtained values corresponds well with discharge behavior via models
- It is evident that increase of gas flow reduces the impurity level

- The model needs further detailed verification
- This will probably be possible only for high ppm values

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Thank you for your attention!