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**Conference or Workshop Item:**

Brisset, Alexandra Helene Marie Brigitte [orcid.org/0000-0003-3217-1106](https://orcid.org/0000-0003-3217-1106), Guichard, F, Cessou, A et al. (1 more author) (2018) Spatio-temporal evolutions of rotational and vibrational temperatures of nitrogen and oxygen of a high field atmospheric discharge in dry air by spontaneous Raman scattering. In: Gordon Research Conference Plasma Processing Science, 05-10 Aug 2018.

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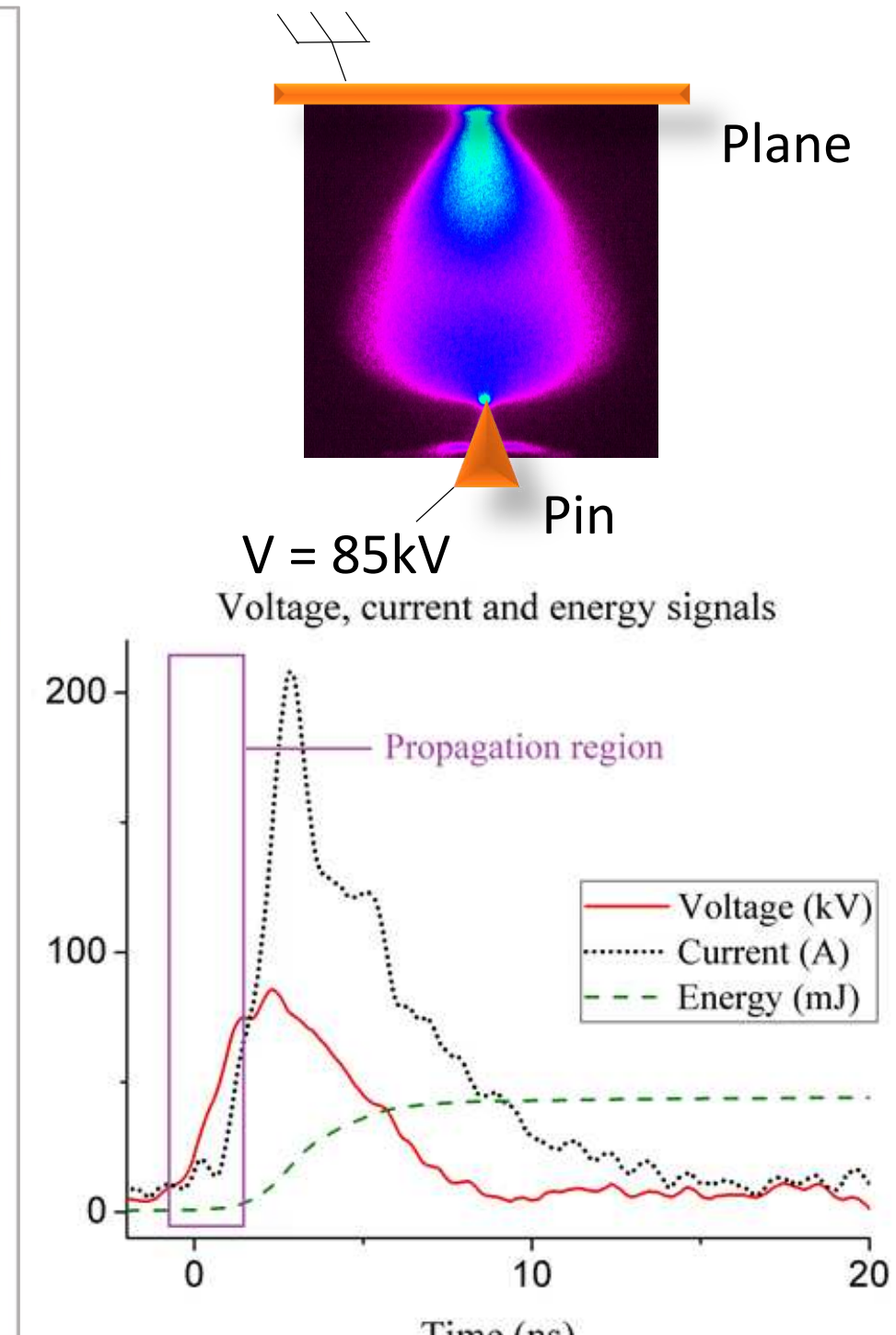
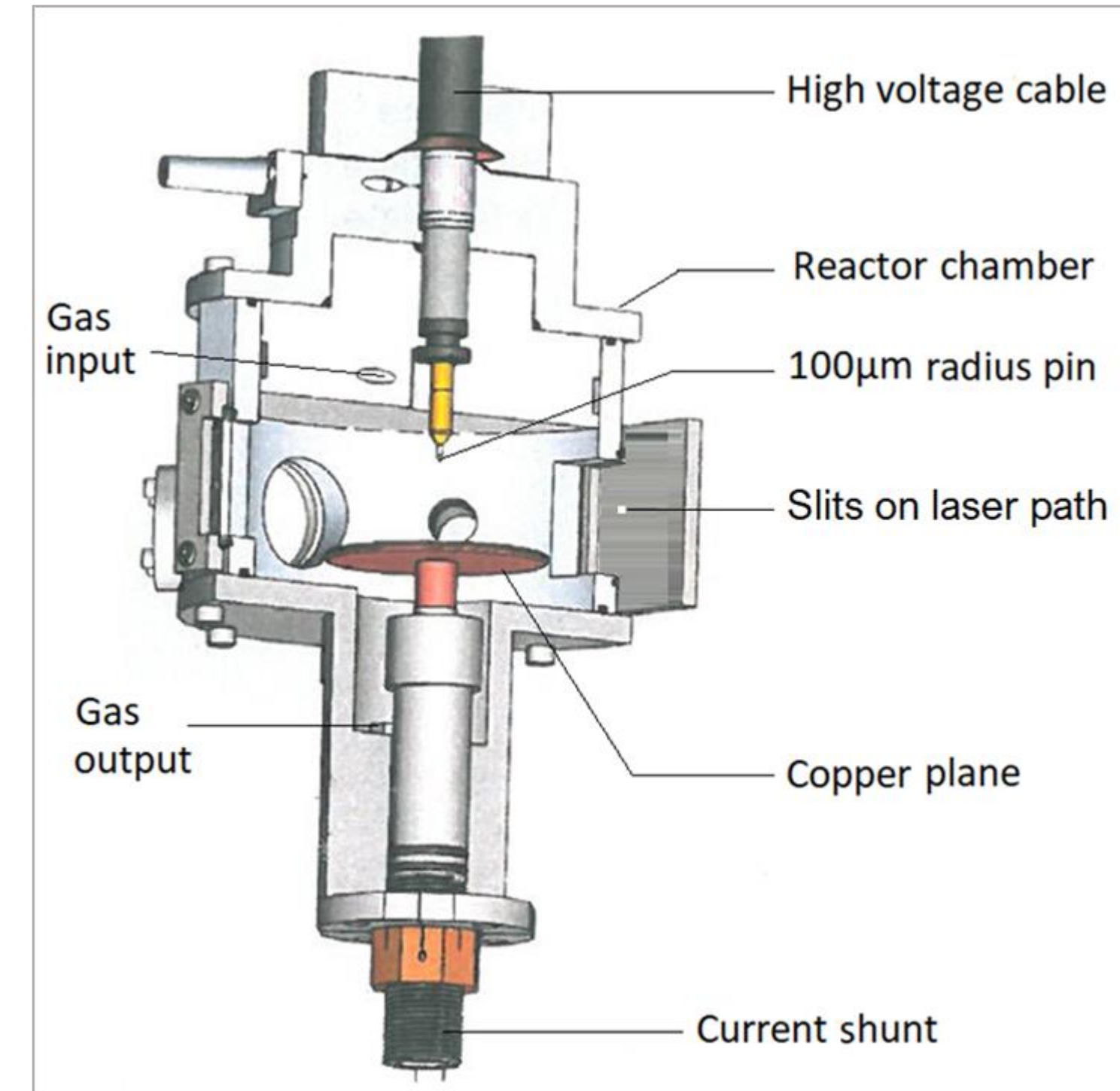
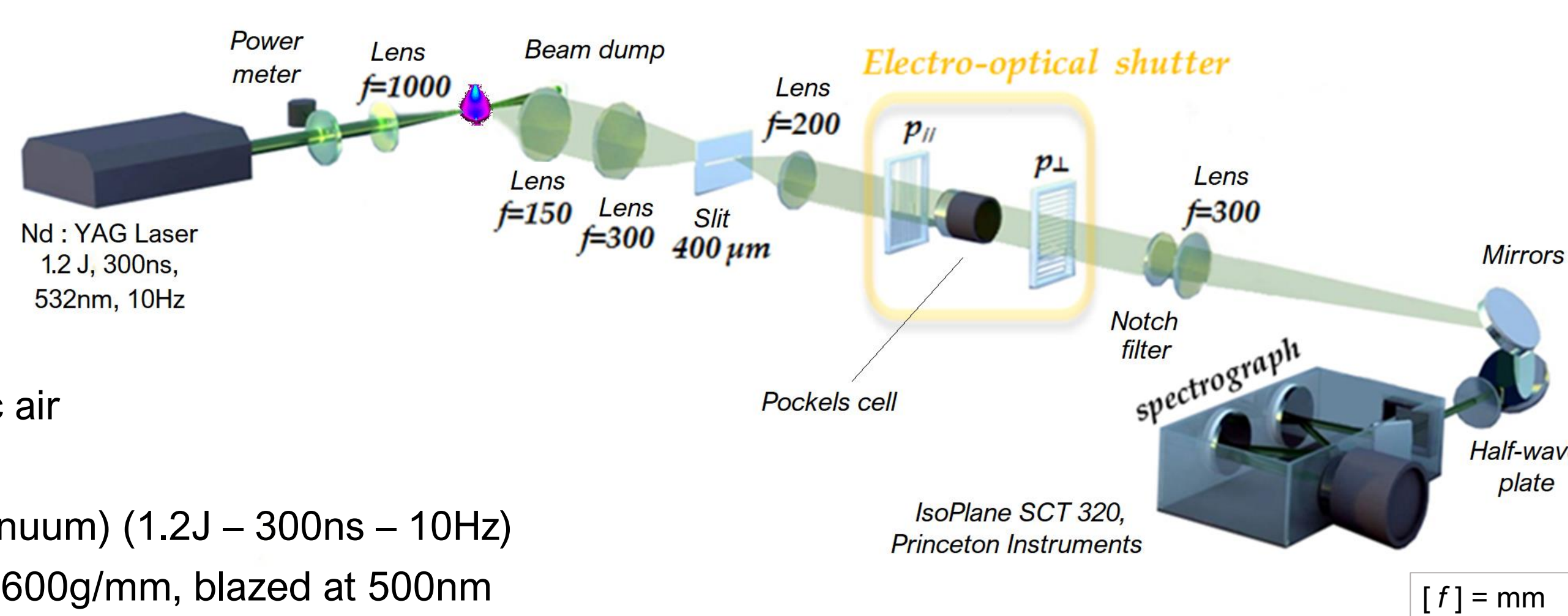
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**Summary:** We report results on the influence of **high electric fields on the energy relaxation and heating mechanisms** of a nanosecond corona discharge in atmospheric air. At 85kV, high non-equilibrium vibrational excitation of nitrogen is observed in the first hundreds of nanoseconds of the post-discharge. **Vibrational-vibrational transfers** dominate until 10 $\mu$ s. Then, **vibrational-translational transfers** participate to **moderate heating of the gas** (about 1000K). **Thermal equilibrium** is reached within 10 to 50ms. At 65kV, energy relaxation time scales are similar but vibrational temperatures are lower and the gas temperature remains below 500K.

**Introduction:** This work comes within a research on the fundamental mechanisms of **diffuse electric discharges under very high electric fields** and the associated non-equilibrium plasmas in atmospheric air. Such discharges are created at very high over-voltages, several tens of kilovolts, with **sub-nanosecond rise times** and without any pre-ionizing system [1]. In the context of air treatment research, better understanding of the physical and chemical processes of these discharges helps to develop advanced and high energy efficiency reactors. The **high field** values that make **large volume discharges**, should enable the design of plasma reactors presenting more homogeneous properties than current ones.

## Experimental setup:

- Pin-plane geometry, 18mm gap
- ~100 $\mu$ m pin radius
- Voltage peaks at 65kV or 85kV
- Repetition frequency: 5Hz
- P=1atm, 1 L.min<sup>-1</sup> flow of synthetic air



- Agilite pulsed Nd:YAG laser (Continuum) (1.2J – 300ns – 10Hz)
- Spectrograph: IsoPlane SCT320 - 600g/mm, blazed at 500nm
- Back-illuminated CCD (Pixis 400B)
- Notch filter at 532nm
- Electro-optical shutter : 2 crossed polarizers + KD\*P Pockels cell (LAP-50, Quantum Tech.)

## Choice of the fitting model according to the state of the gas [2]:

Various energy distribution functions have to be considered to correctly describe reactive media because of strong energy transfers.

- A **model at thermodynamic equilibrium** for spatial positions out of the discharge and all positions after complete energy relaxation:

$$N_{v,J} = N \frac{(2J+1)g_J \exp\left(-\frac{hcE_{vib}(v)}{kT} - \frac{hcE_{rot}(v,J)}{kT}\right)}{Q_{vib}(T)Q_{rot}(v,T)} \quad T_{trans} = T_{rot} = T_{vib}$$

- A **three temperatures model** for times before vibrational relaxation:

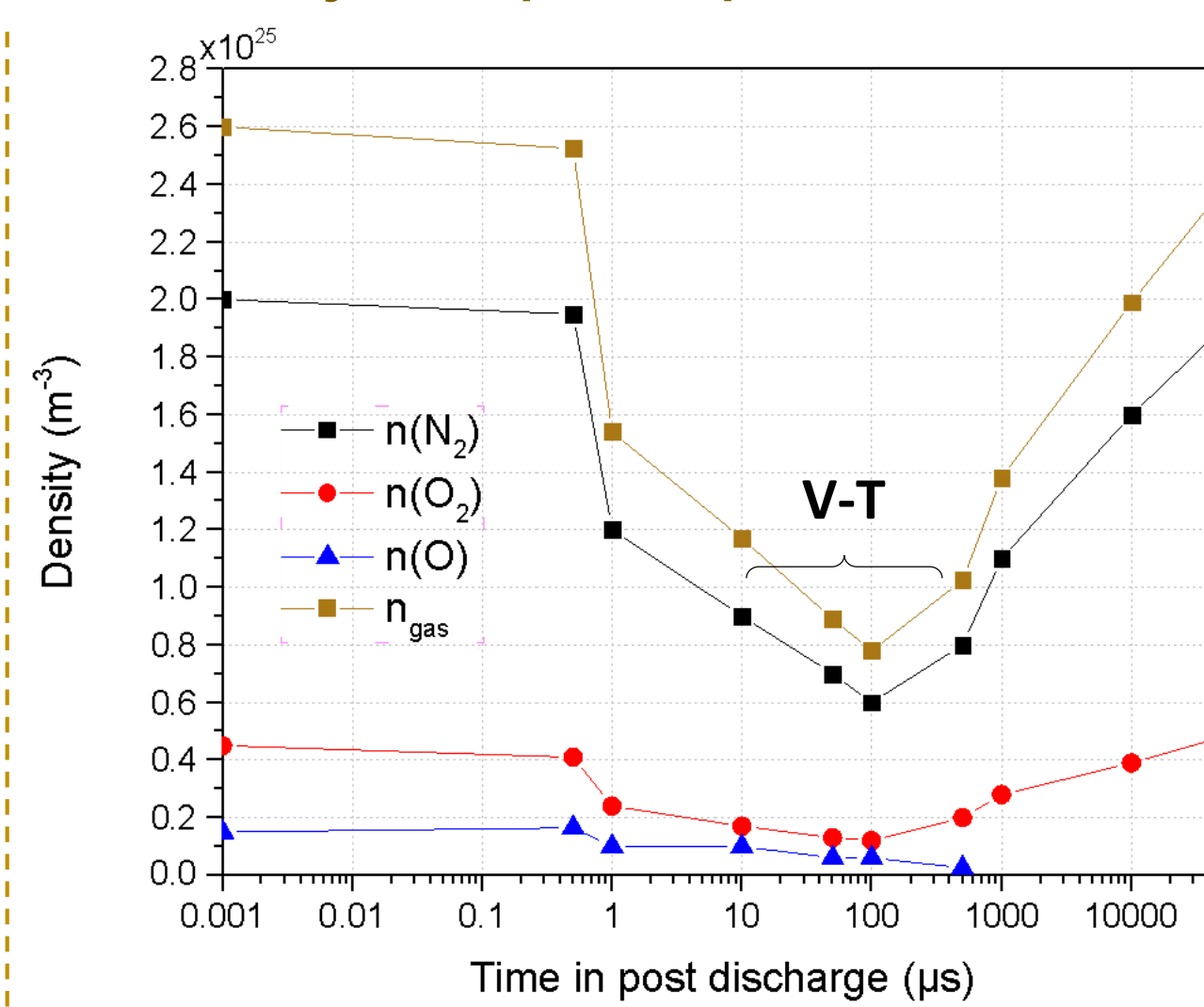
$$N_{v=0,J} = N \frac{(2J+1)g_J \exp\left(-\frac{hc(E_{vib}(v=1) - E_{vib}(v=0))}{kT_{v01}} - \frac{hcE_{rot}(v=1)}{kT_{v1v}} - \frac{hcE_{rot}(0,J)}{kT_{rot}}\right)}{Q_{vib}(T_{v01}, T_{v1v})Q_{rot}(0, T_{rot})}$$

$$N_{v>0,J} = N \frac{(2J+1)g_J \exp\left(-\frac{hcE_{vib}(v)}{kT_{v1v}} - \frac{hcE_{rot}(v,J)}{kT_{rot}}\right)}{Q_{vib}(T_{v01}, T_{v1v})Q_{rot}(v, T_{rot})} \quad T_{rot} \neq T_{v=0,1} \neq T_{v>1}$$

- A **two temperatures model** for times after vibrational relaxation:

$$N_{v,J} = N \frac{(2J+1)g_J \exp\left(-\frac{hcE_{vib}(v)}{kT_v} - \frac{hcE_{rot}(v,J)}{kT_{rot}}\right)}{Q_{vib}(T_v)Q_{rot}(v, T_{rot})} \quad T_{rot} \neq T_{vib}$$

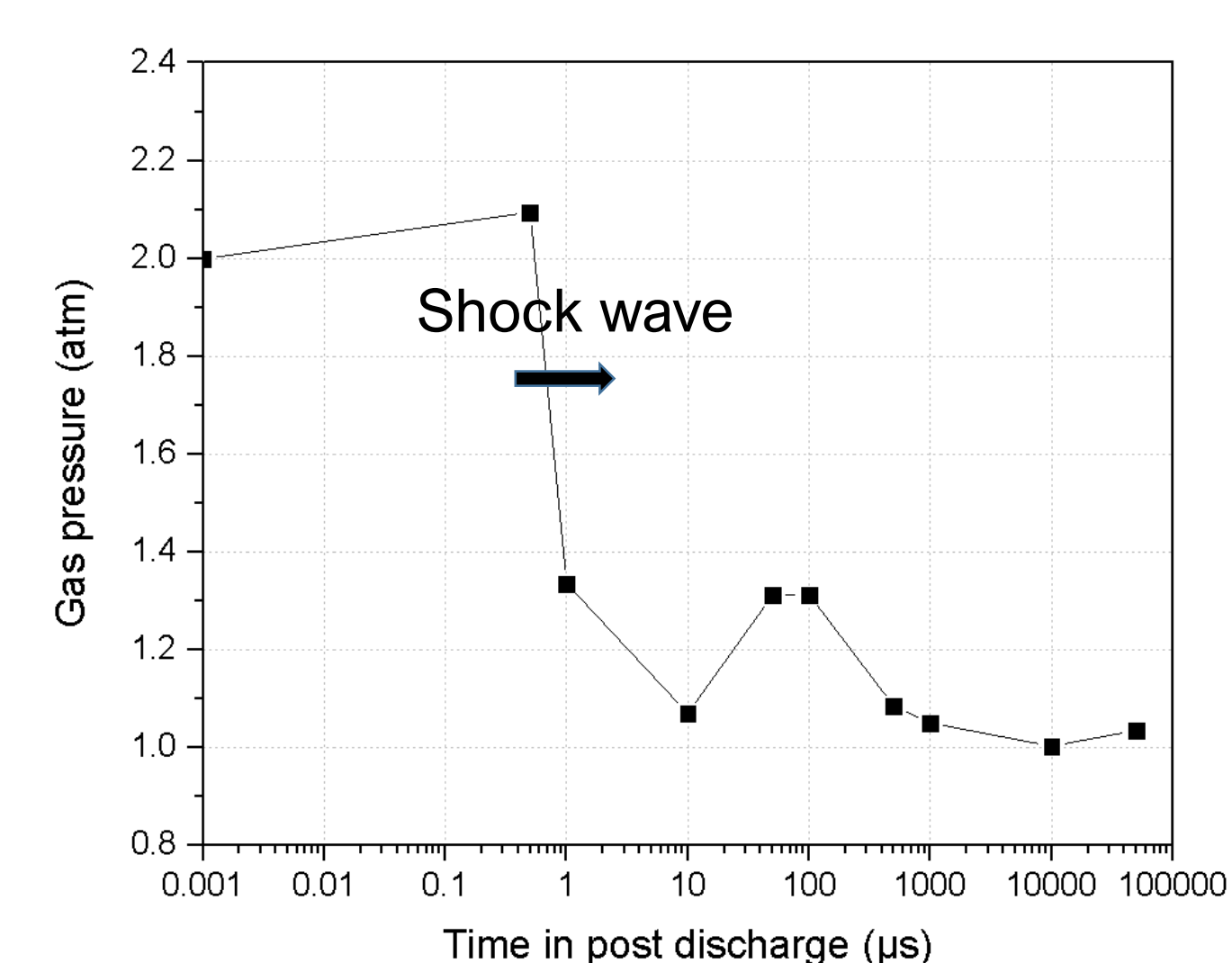
## Density temporal profiles:



- Just after the discharge:**
- Isochoric heating of the gas
  - Significant dissociation of oxygen molecules
  - Gas expansion starts

- At 1 $\mu$ s:**
- Shock wave re-establishes pressure

## Pressure temporal profiles:



- From 10 $\mu$ s to 500 $\mu$ s:**
- Enhanced gas expansion with heating due to V-T transfers
- From 500 $\mu$ s to 10ms:**
- Relaxation by diffusion

## O<sub>2</sub> dissociation model:

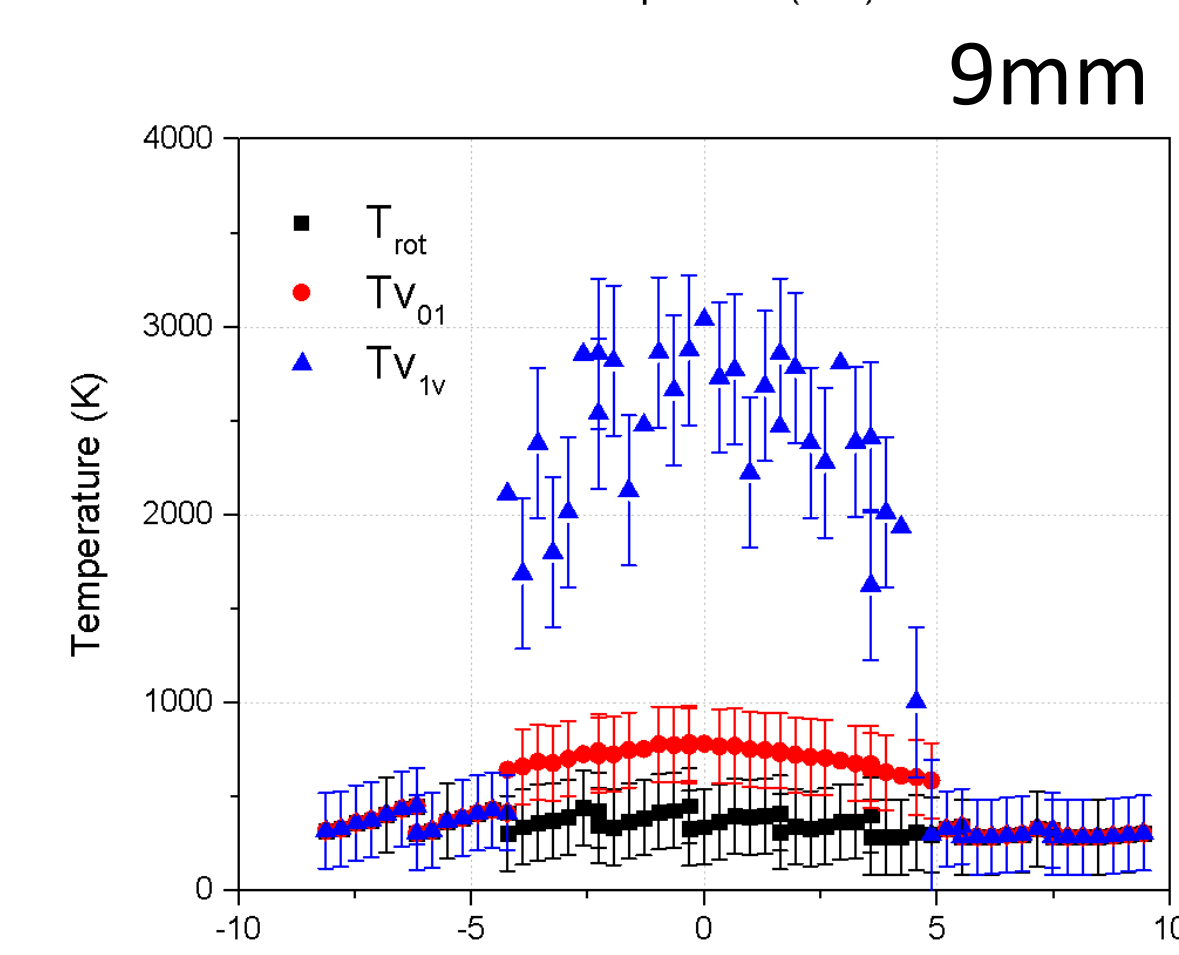
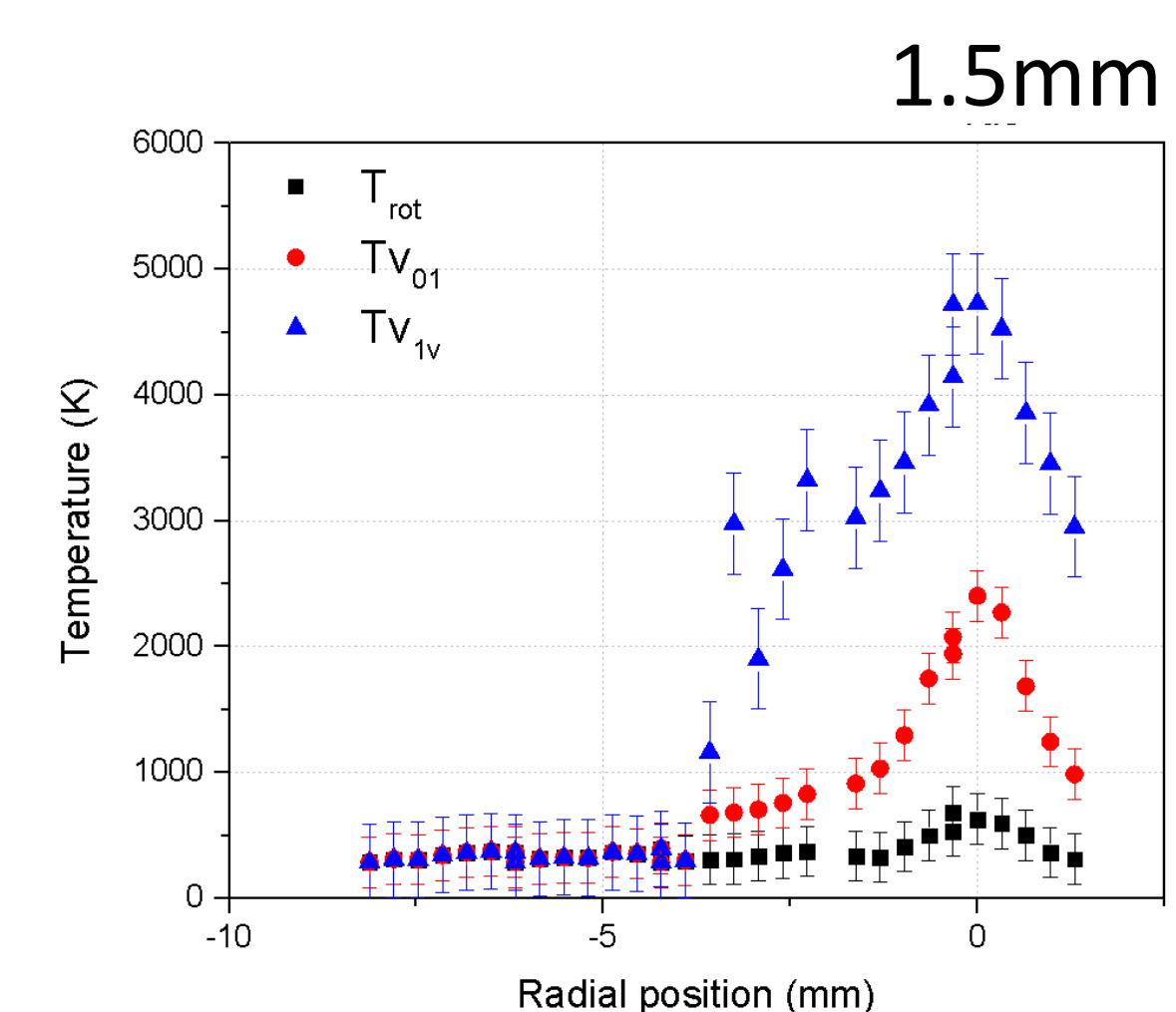
$n(O)$  is determined with the evolution of the ratio of O<sub>2</sub> and N<sub>2</sub> atoms compared with the equilibrium ratio 1:4

$$\tau_{dissociation}(O_2) = \frac{1/2n(O)}{n(O_2) + 1/2n(O)}$$

## 85kV – dry air – at the pin :

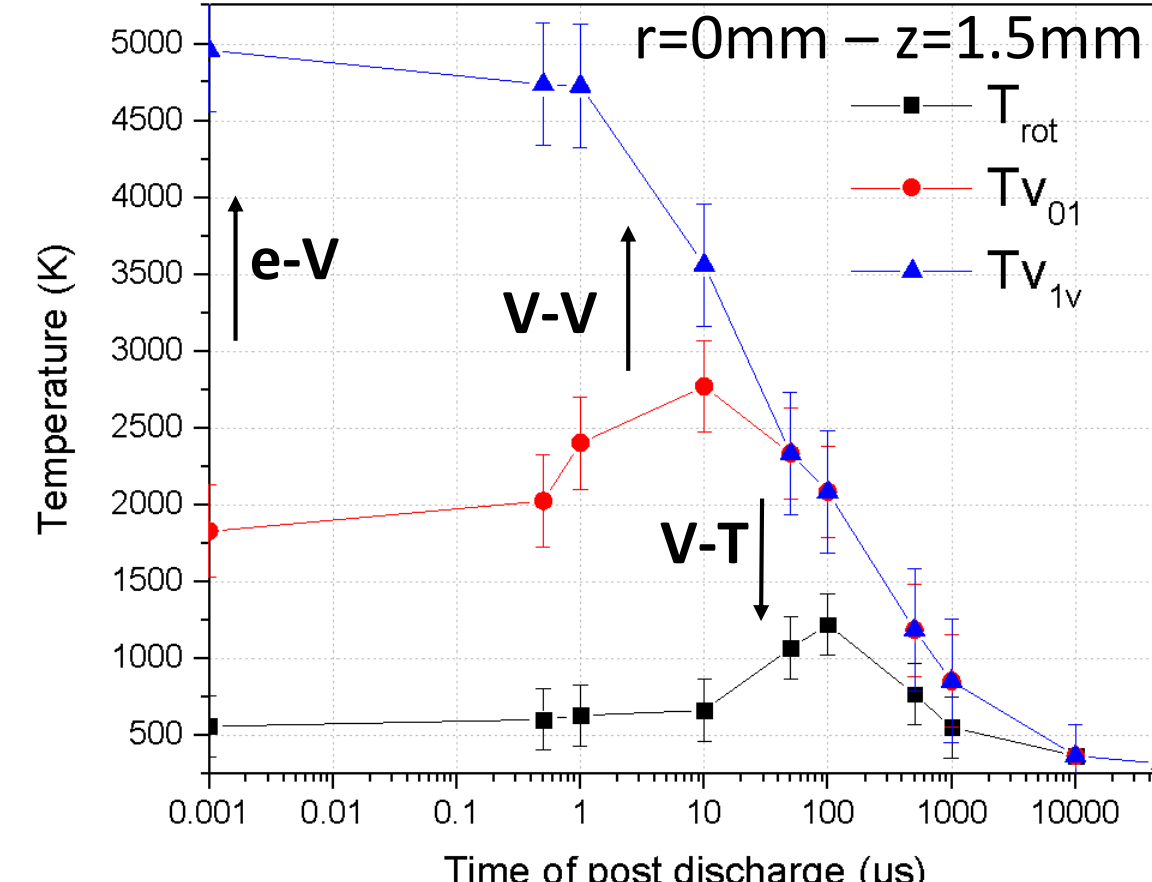
- Just after the discharge, ~15% of O<sub>2</sub> are dissociated.
- Density of O atoms is divided by two in 100 $\mu$ s.
- O atoms are recombined in 500 $\mu$ s.

## Radial profiles at 85kV – 1 $\mu$ s:

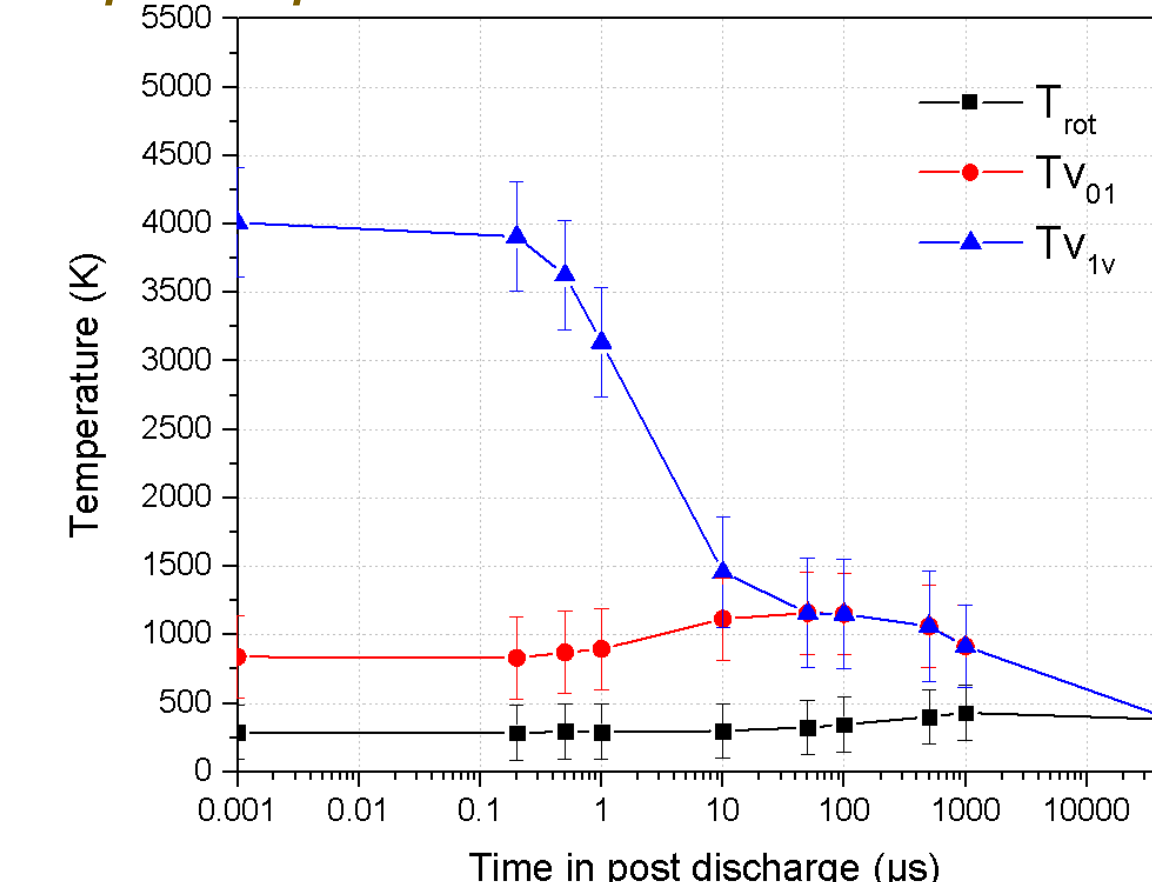


- **Radial temperature extent** depends on the vibrational level considered.
- The radial extent is **coherent with the light emission profiles**.
- The rotational temperature can only be determined with **good sensitivity over 500K**. Below, it is influenced by the accuracy of the instrumental function for each region of interest.

## Temporal profiles at 85kV: 1.5mm



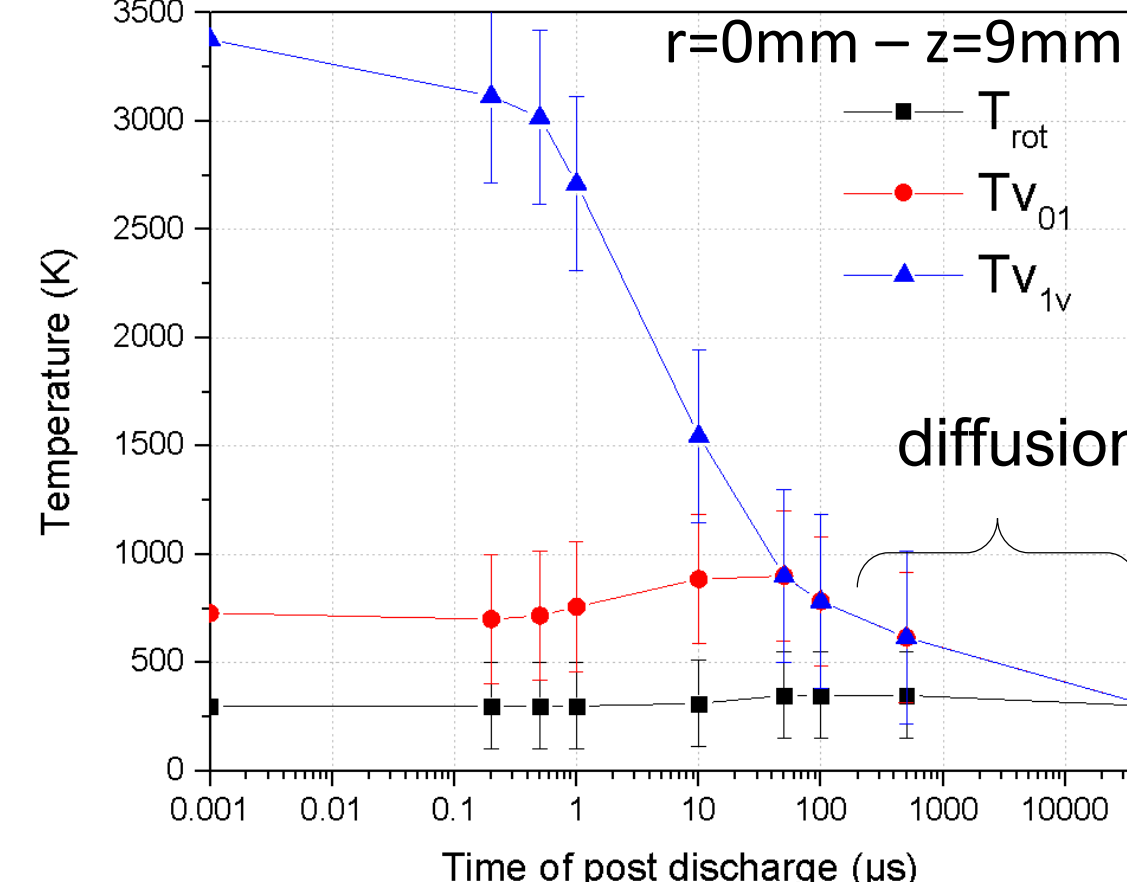
## Temporal profiles at 65kV: 1.5mm



### At 1.5mm:

- **Gas heating requires strong non-equilibrium** induced by very high voltages.
- Voltage impact on vibrational non-equilibrium and induced V-T gas heating processes is mostly **localised at the pin**.

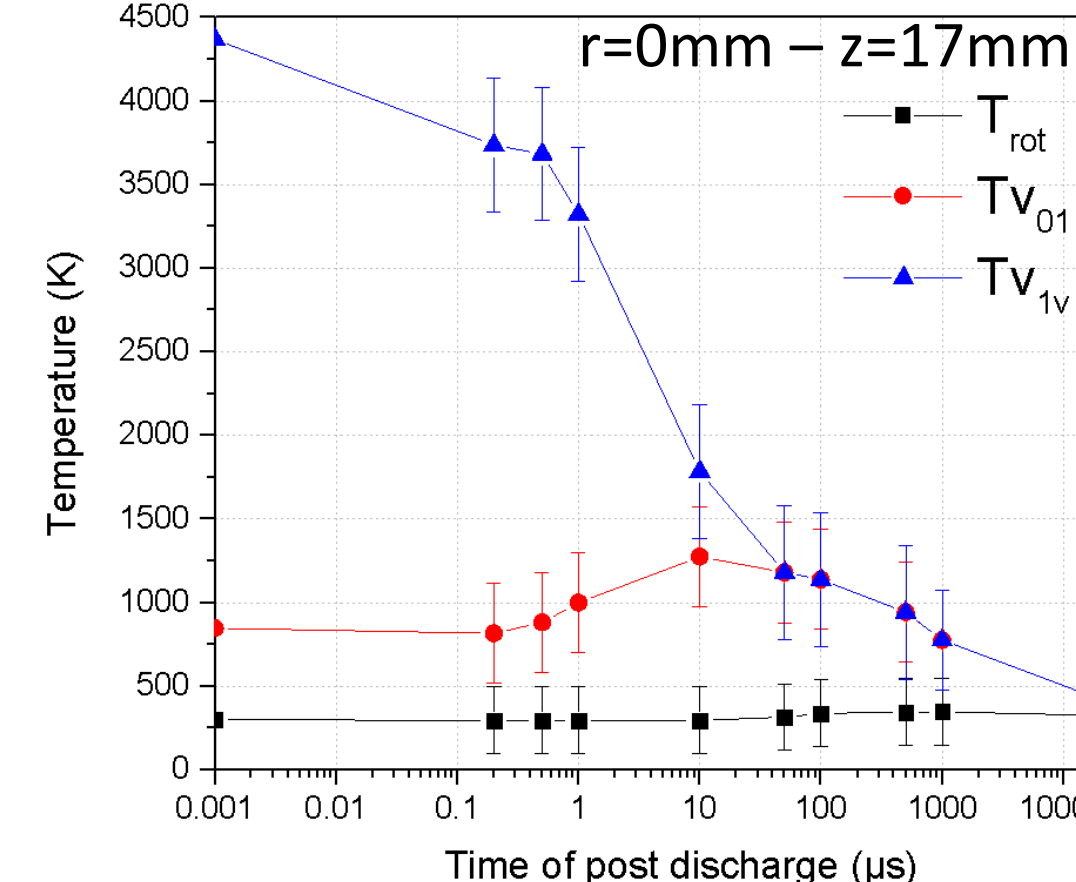
## 9mm



### At 9mm:

- **Close to equilibrium** between vibrational and rotational levels at low voltage.
- Excitation of  $v=0,1$  **under detector sensitivity at 65kV**.
- **Almost no heating** of the gas in most of the discharge volume up to 85kV.

## 17mm



### At 17mm:

- **Low effect of voltage** between 65kV and 85kV.
- Gas heating at the limit of detection sensitivity.

- ❖ Relaxation time scales are similar whatever the position and voltage. The same mechanisms prevail.

### Just after the discharge:

- **e-V transfers** during the discharge => high non-equilibrium vibrational distribution.
- **Fast heating** dominantly due to [3]:
  - Quenching of N<sub>2</sub>(C,B) states by O<sub>2</sub>
  - Dissociation of O<sub>2</sub> and N<sub>2</sub> by electronic impact
  - Quenching of O(1D) with N<sub>2</sub> and O<sub>2</sub>
  - Reactions involving charged particles

### Up to 10 $\mu$ s:

- **V-V transfers** dominate from high to low vibrational levels
- Vibrational equilibrium is reached between **10 and 50 $\mu$ s**
- Low V-T transfers
- Ongoing chemical heating balanced by adiabatic cooling → +100K at the pin

### 10 $\mu$ s to 500 $\mu$ s:

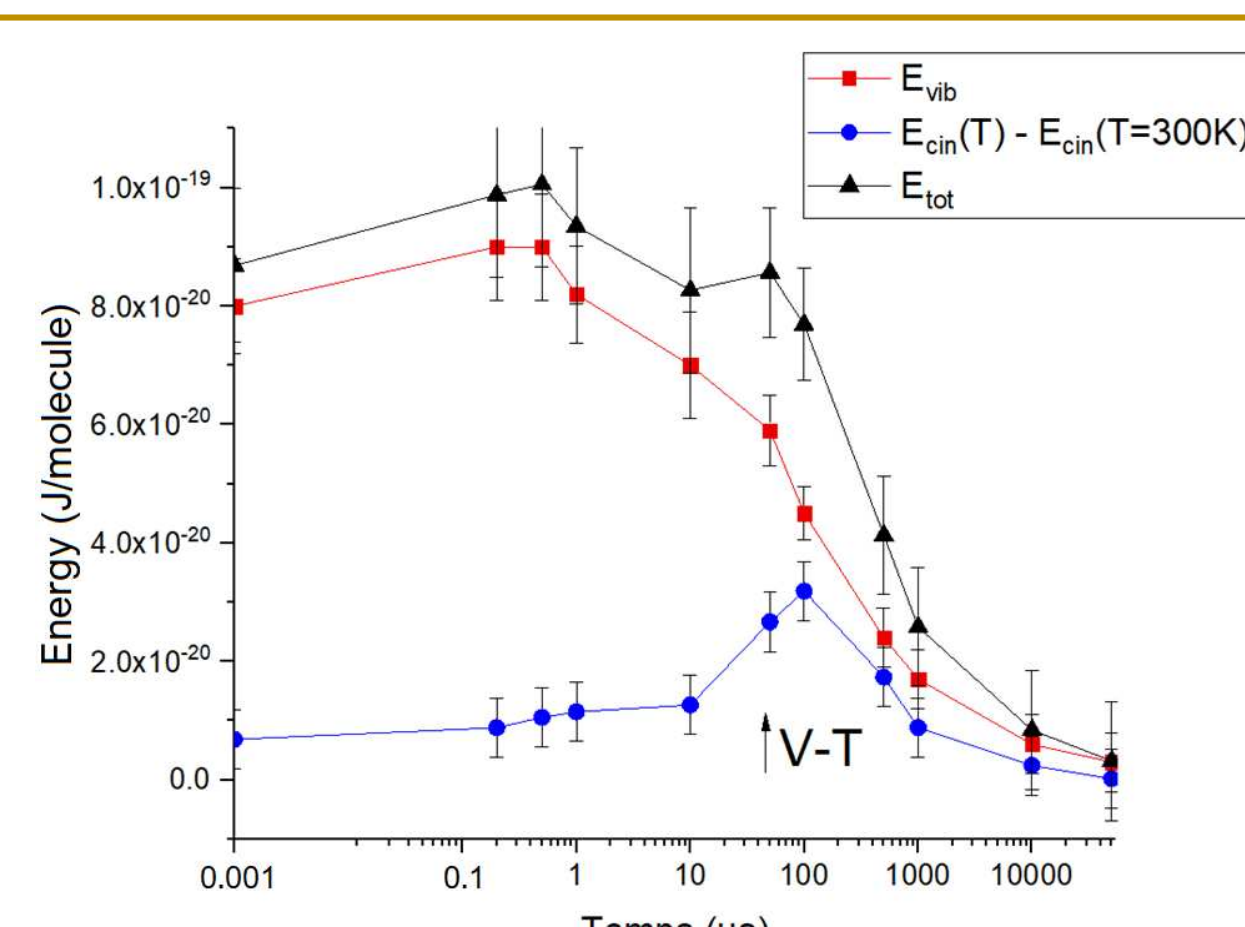
- V-T transfers dominate: gas heating

### 500 $\mu$ s to 1ms:

- Cooling by gas diffusion and convection

## Energy per molecule of N<sub>2</sub>(X):

- ~90% E<sub>vib</sub> at t=100ns
- E<sub>cin</sub> constant over 10 $\mu$ s: low V-T transfers
- E<sub>vib</sub> decrease slowly from 1 $\mu$ s: choc wave
- Heating: V-T transfers
- E<sub>vib</sub> fall, E<sub>cin</sub> fall: relaxation by diffusion



**Conclusion:** Highly non-equilibrium discharge shows ~90% of the energy of N<sub>2</sub> molecules is stored in vibrational excitation at the end of the discharge and up to ~15% of O<sub>2</sub> molecules are dissociated. In dry air, fast heating is low and very localised at the pin. A shock wave re-establishes pressure at 1 $\mu$ s. V-V transfers dominate from 1 to 50 $\mu$ s, followed by V-T transfers. Significant induced heating is localised at the pin and reaches no more than 1000K and requires high voltages. Voltage impacts the energy share between vibrational levels: excitation of levels 0 and 1 in the volume requires voltages >65kV. High temperatures maintained over more than 100 $\mu$ s on mm scale regions at the pin can compete with spark discharges for combustion and high vibrational excitation on large radial extent promotes reactivity.

## References:

- [1] P. Tardiveau, L. Magne, E. Marode, K. Ouaras, P. Jeanney and B. Bournonville, Plasma Sources Sci. Technol. 00 (2016) 000000 (16pp)
- [2] A. Lo, G. Cléon, P. Vervisch, A. Cessou, J. Phys. B: Appl. Phys. 107 229 (2012)
- [3] N. A. Popov, J. Phys. D: Appl. Phys. 44 285201 (2011)

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