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Vibrational Excitation of CO₂ by Nanosecond Discharges at Atmospheric Pressure

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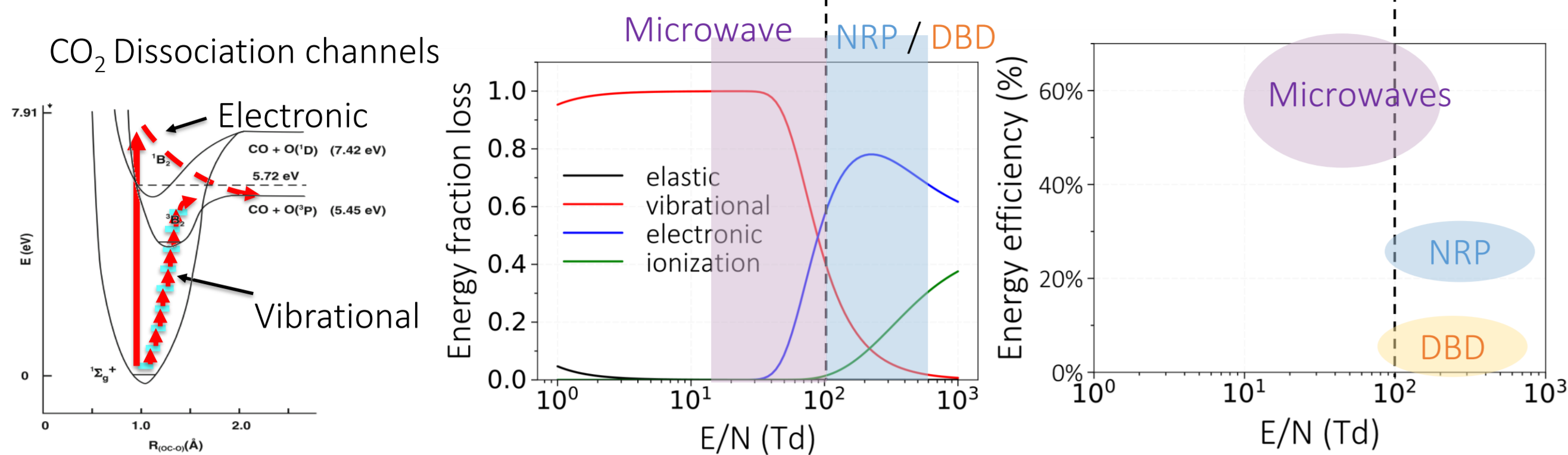
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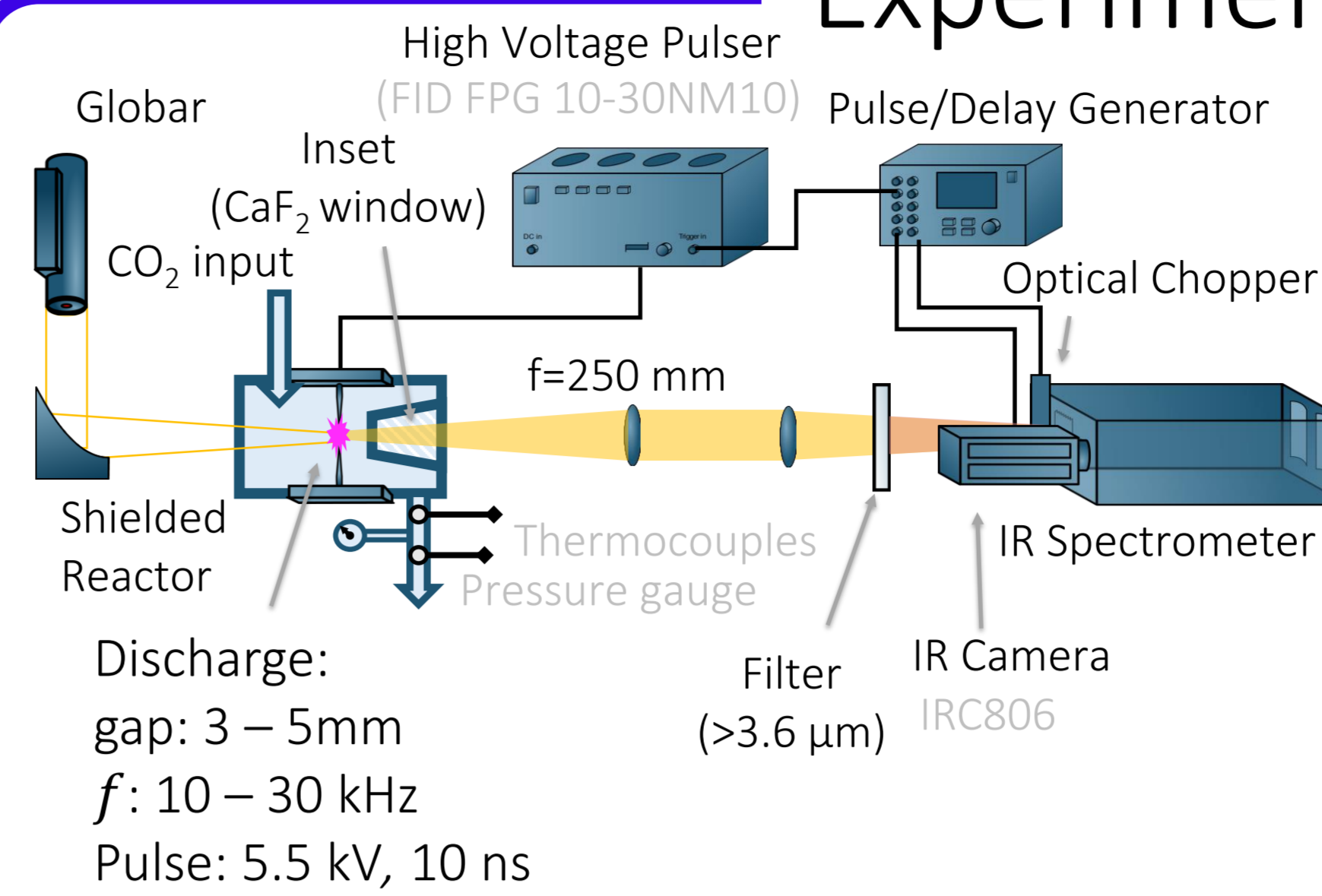
Motivation

CO₂ dissociation efficiency is maximized with high vibrational excitation (T_{vib}) and low heating (T_{trans}) [1]. Nanosecond Repetitive Pulses (NRP) are more efficient than DBD [2] at similar reduced field (E/N). Why?



Goal: **measure the vibrational contribution in NRP sparks CO₂ splitting**
How: OES meas. + Line by line (LBL) model + Line-of-sight (LOS) model

Experimental Setup



Measurements:

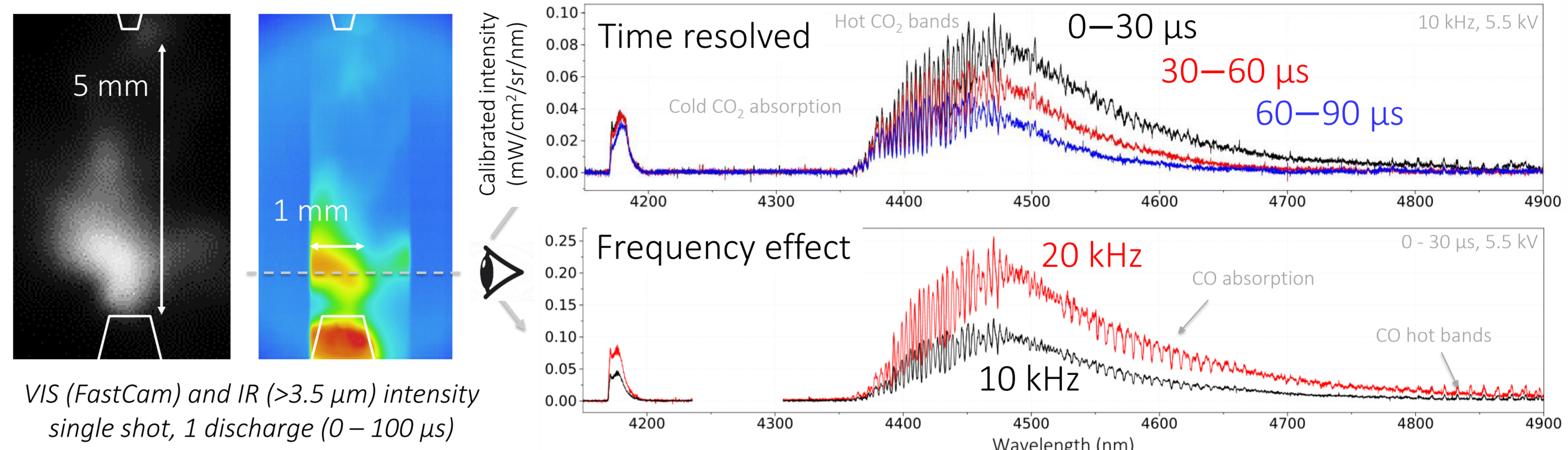
- CO₂: T_{vib} , T_{rot} , N
- mid-IR Absolute optical emission spectroscopy (OES), time-resolved (10 μ s), spatially resolved (500 μ m)
- CO: T_{vib} , T_{rot} , N
- Broadband IR Absorption production in post-discharge

Alternative techniques:
 - CO₂ Raman scattering: difficult to analyze ✗
 - FTIR: excellent time, spectral resolution [3] ✓ but no spatial resolution ✗

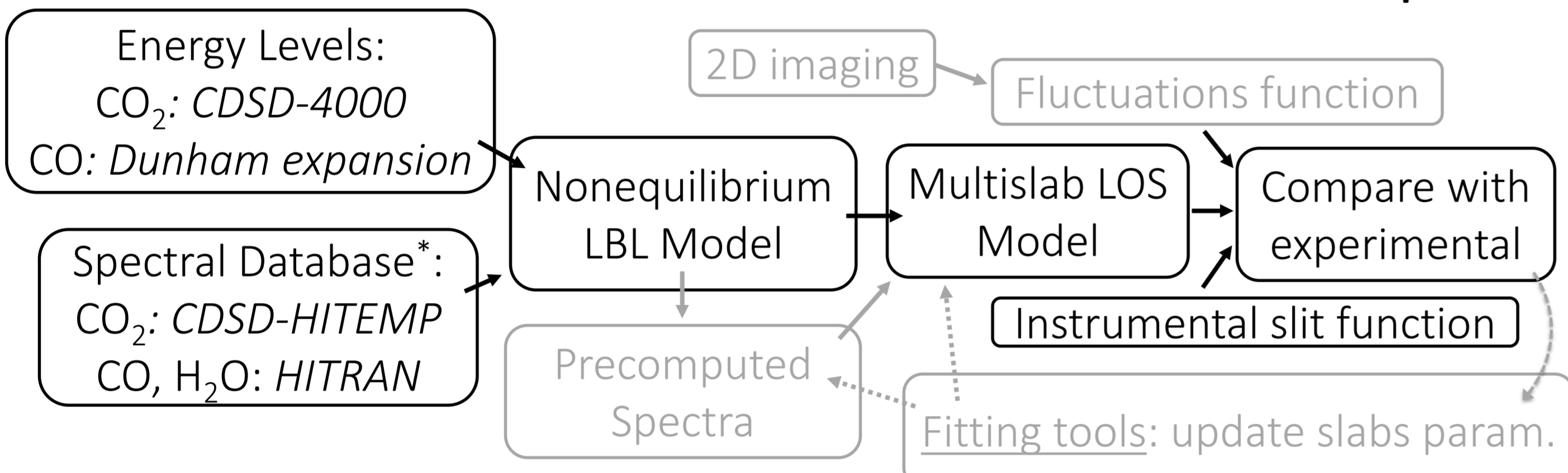
Challenges

- Electromagnetic interferences → shielded reactor design ✓
- Room air absorption → optical train purged for absolute calibration ✓
- Spatial fluctuations (hydrodynamics) → spatially resolved spectra ✓
- Signal-to-dark ratio ~ 1/400 → phase locked capture of dark drift ✓
- Camera mistrigs → post-processing detection algorithm ✓
- 2D: Large data generation (~ 8 hrs, 200 Gb / full spectrum)
- Atmospheric pressure: broad, overlapping lines

Data

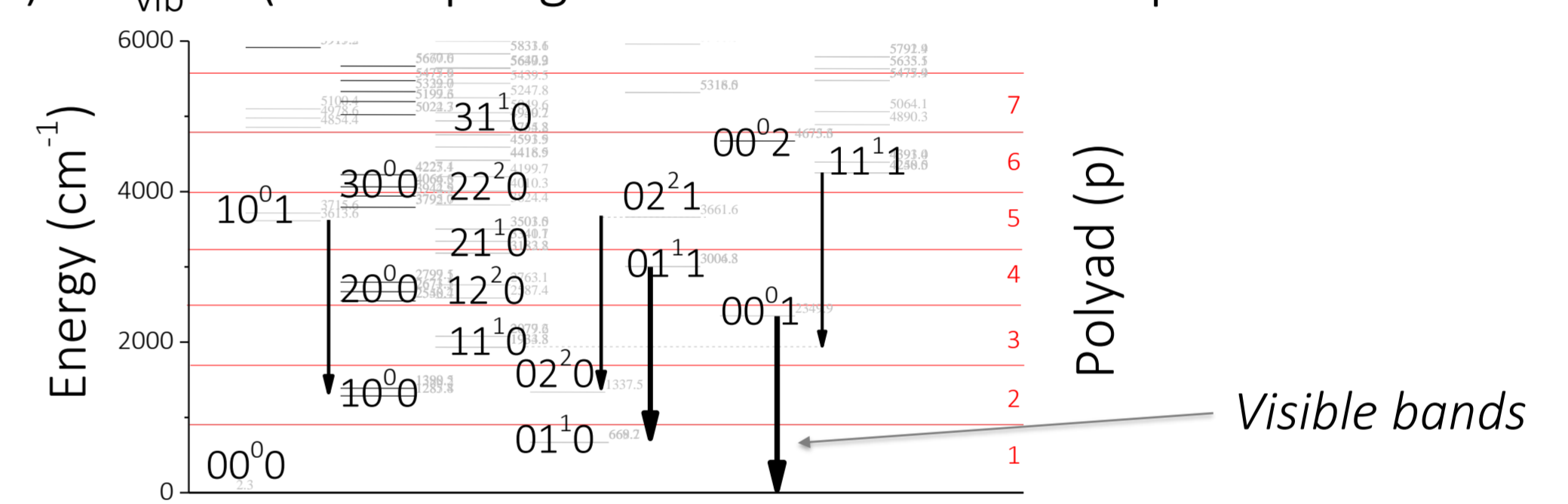


Nonequilibrium LBL Model



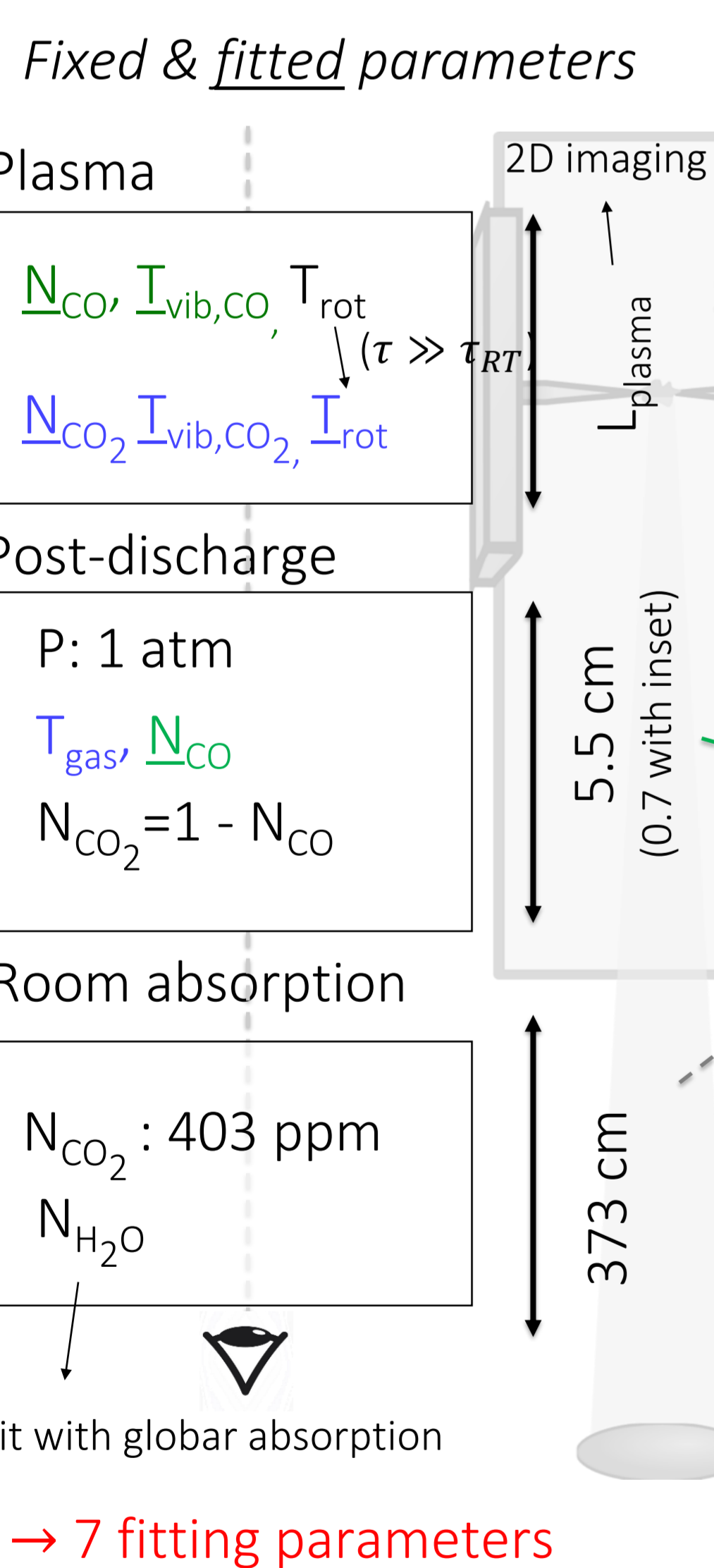
CO₂ vib / rot coupling: $E = E_{vib} + E_{rot} + E_{coupling}$. In CDS polyad nomenclature (p,J,c,N) we chose $E_{vib} = E(p,0,c,N)$, $E_{rot} = E(p,J,c,N) - E_{vib}$ (→ coupling included in rotational partition function)

CO₂ low lying level (HITRAN notation vs polyads)

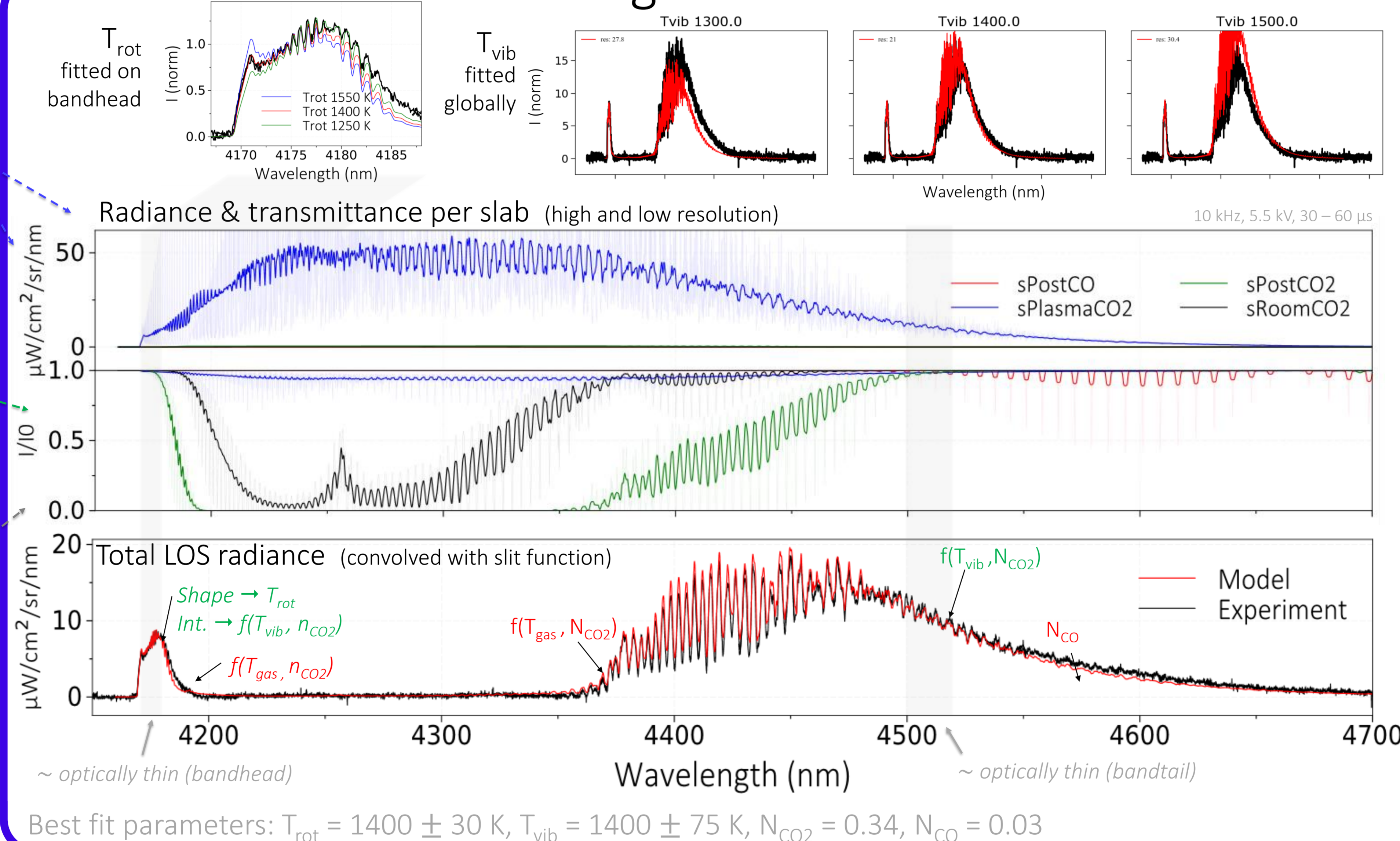


Distribution: overpopulation factors allow arbitrary non equilibrium distributions of vibrational levels; rotational ~ Boltzman

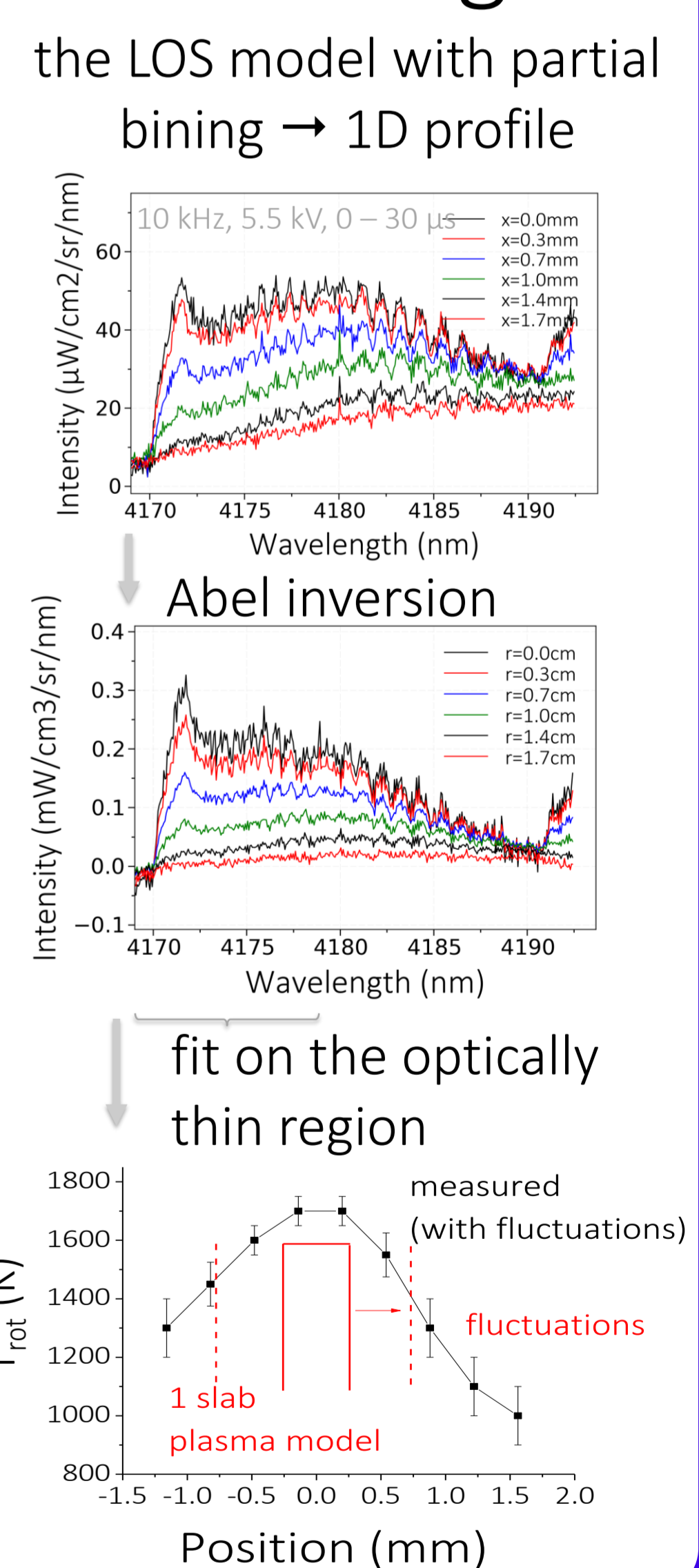
LOS Model



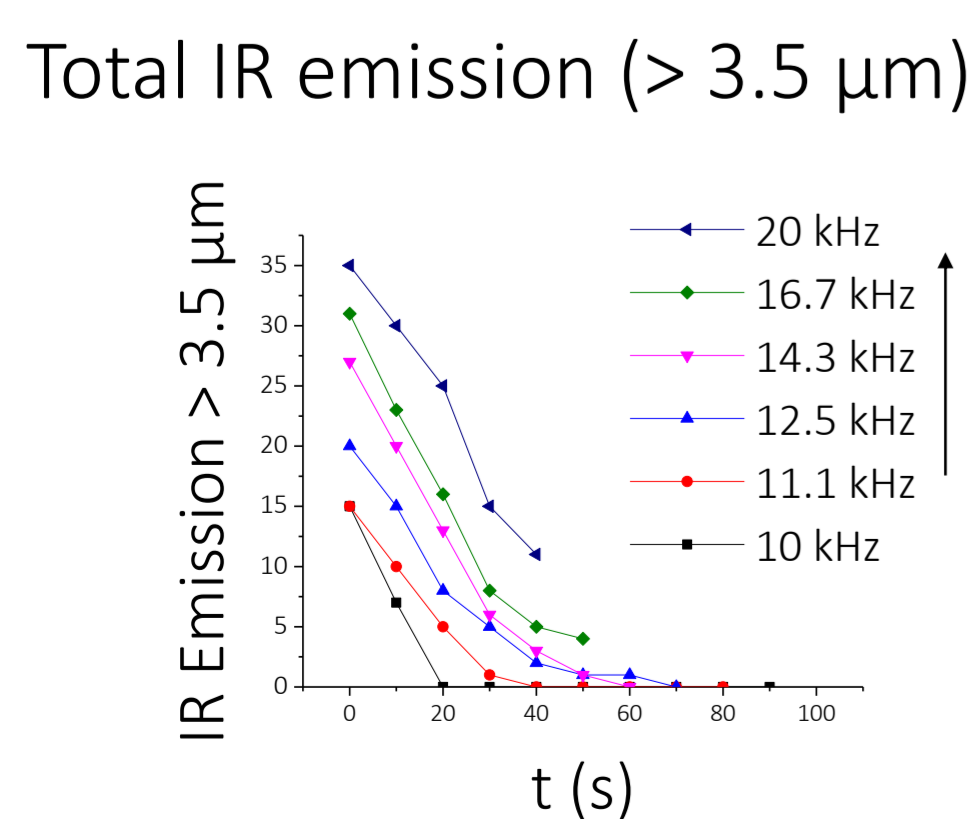
Fitting method



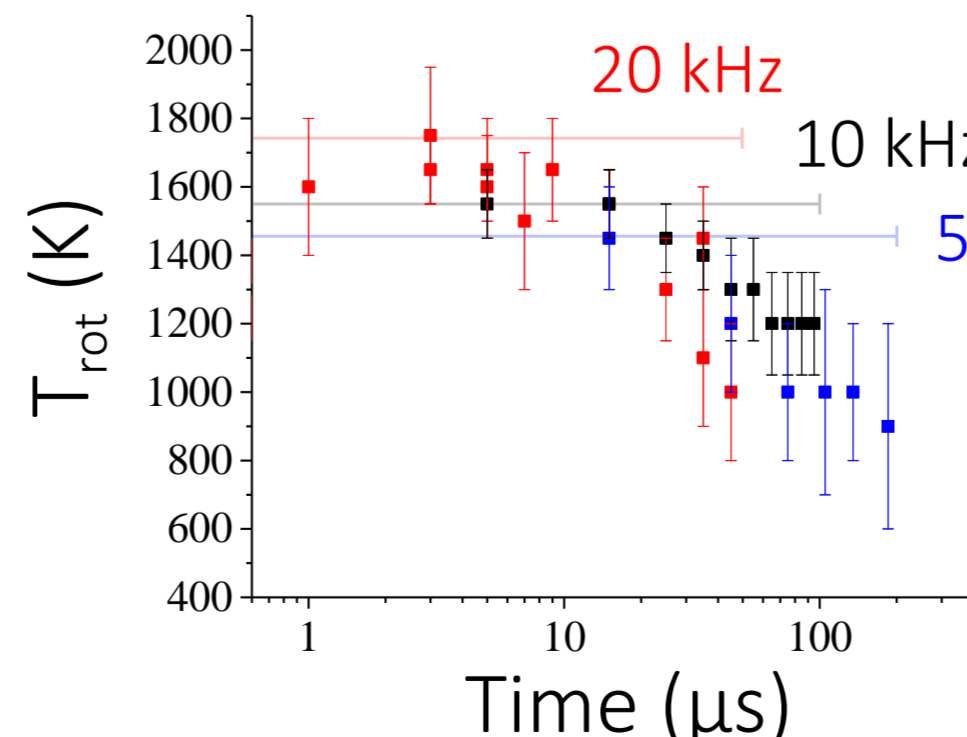
Refining



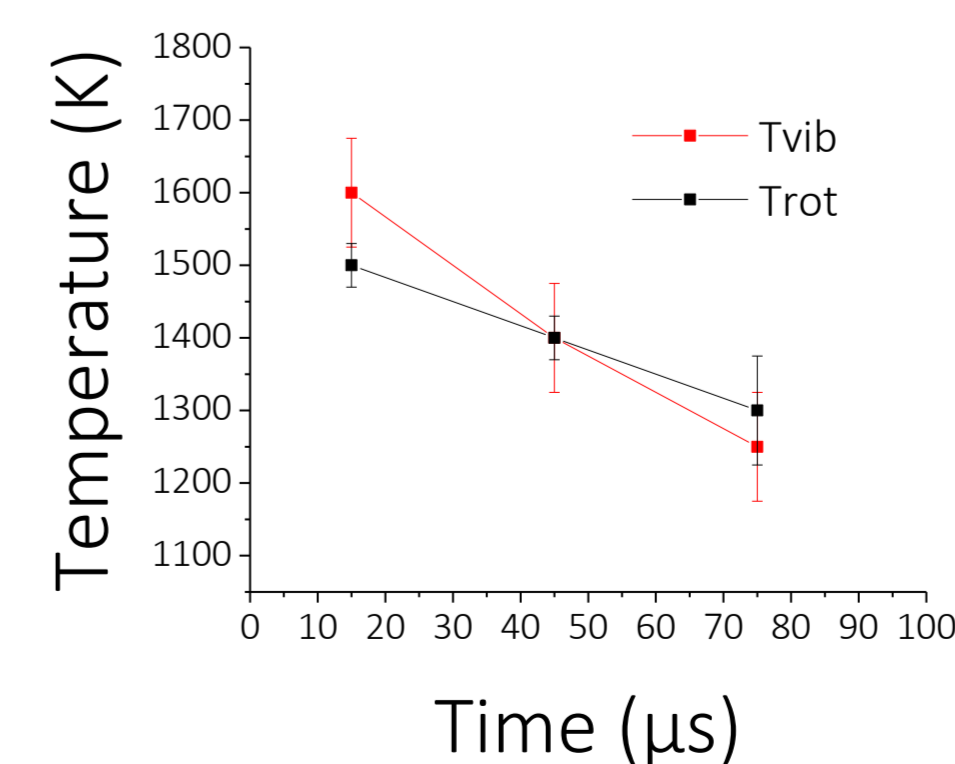
Results & Conclusions



T_{rot} measured by bandhead fitting (2, 10, 30 μ s period)



T_{vib} measured by global fitting (30 μ s period, 10 kHz)



Fitting results:

- 10 kHz: $T_{vib} - T_{rot}$ equilibrium achieved within the first 30 μ s after the discharge. Correct modelling can be obtained with a simplified 3-slabs model
- 20 kHz: evidence of non-equilibrium in the first 30 μ s with a departure from a 1- T_{vib} Boltzmann vibrational distribution model

Next step: compare measured vibrational populations with predictions of a 0D-kinetic model

