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Vibrational Excitation of CO₂ by Nanosecond Discharges at Atmospheric Pressure

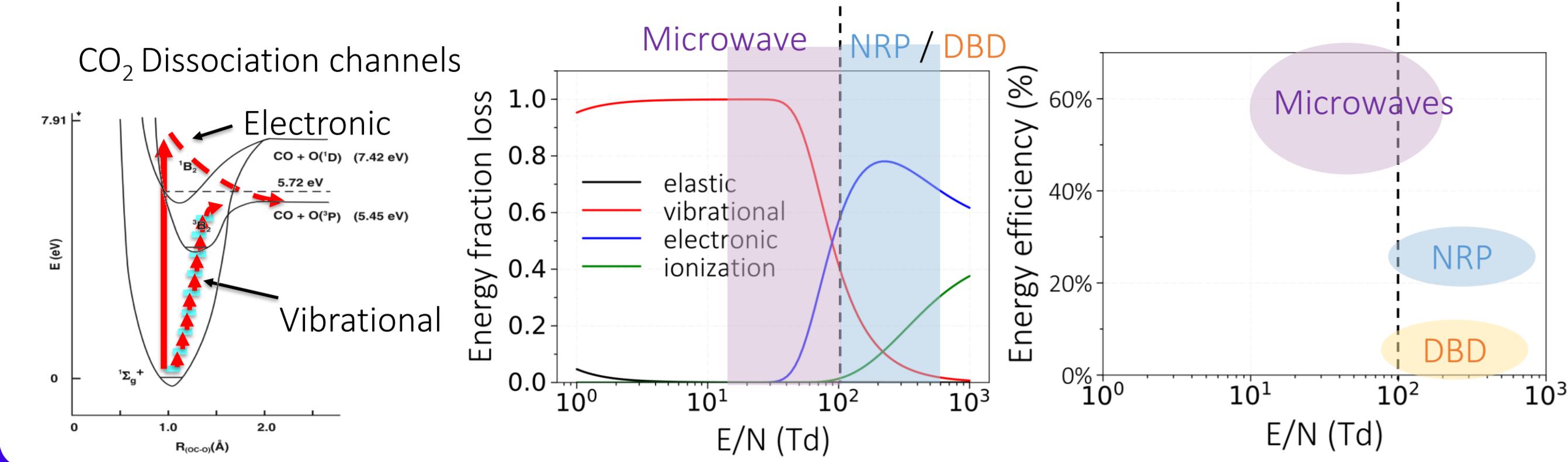
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Motivation

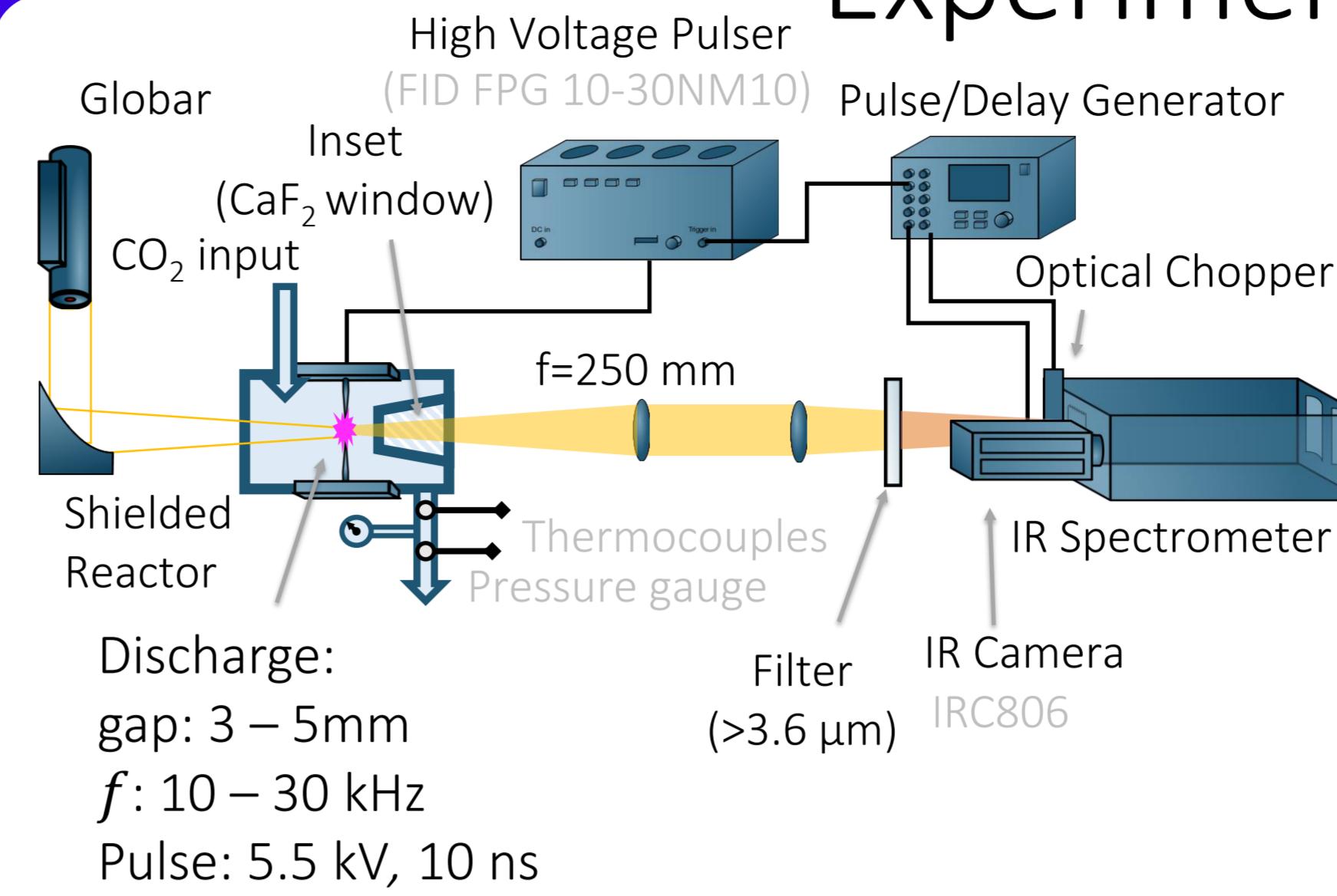
CO₂ dissociation efficiency is maximized with high vibrational excitation (T_{vib}) and low heating (T_{trans}) [1]. Nanosecond Repetitive Pulses (NRP) are more efficient than DBD [2] at similar reduced field (E/N). Why?



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 μs), spatially resolved (500 μm)

CO: T_{vib} , T_{rot} , N

- Broadband IR Absorption production in post-discharge

Alternative techniques:

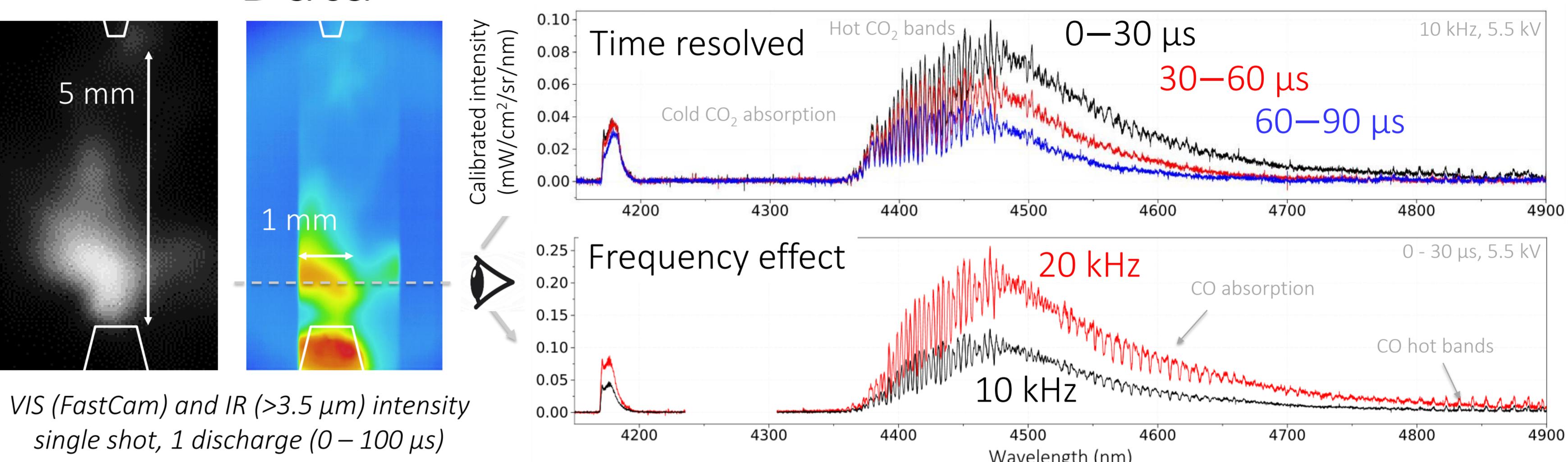
- CO₂ Raman scattering: difficult to analyze ✗

- FTIR: excellent time, spectral resolution [3] ✓ but no spatial resolution ✗

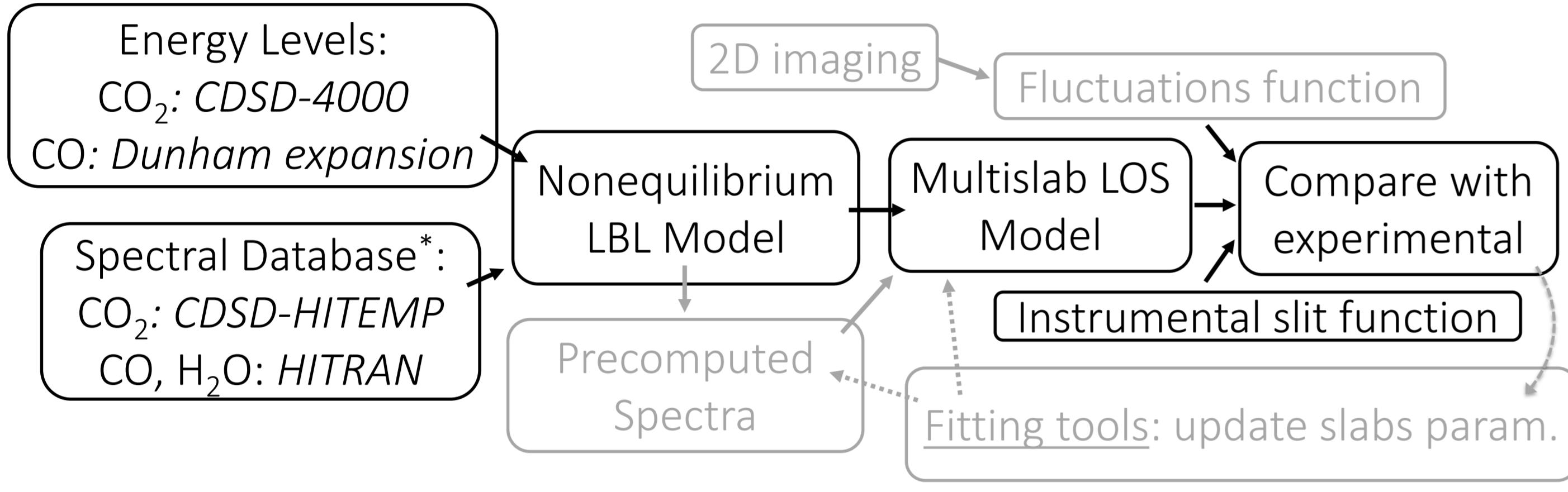
Challenges

- 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 mists → post-processing detection algorithm ✓
- 2D: Large data generation (~ 8 hrs, 200 Gb / full spectrum)
- Atmospheric pressure: broad, overlapping lines

Data

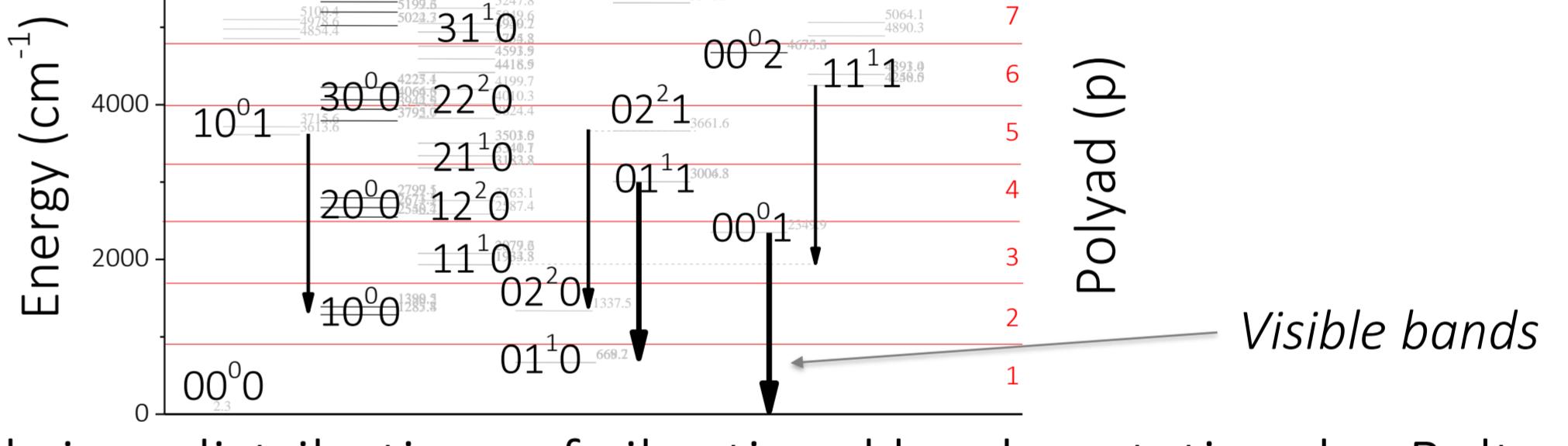


Nonequilibrium LBL Model



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

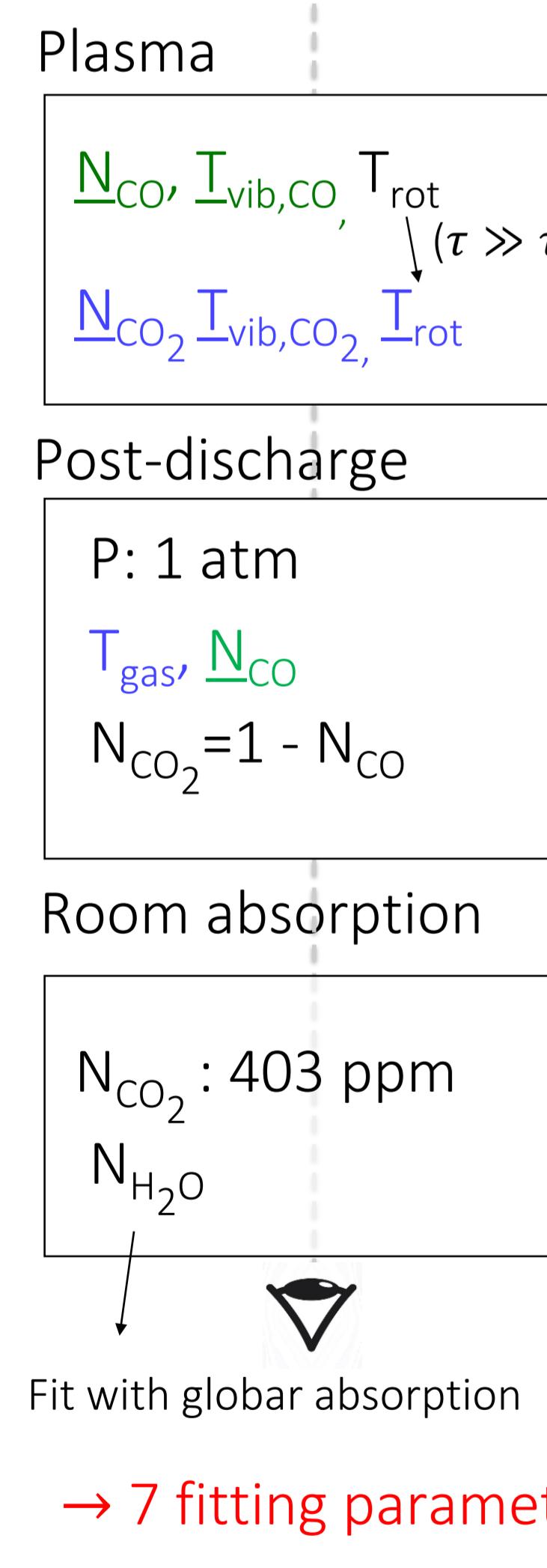
CO₂ low lying level (HITRAN notation vs polyads)



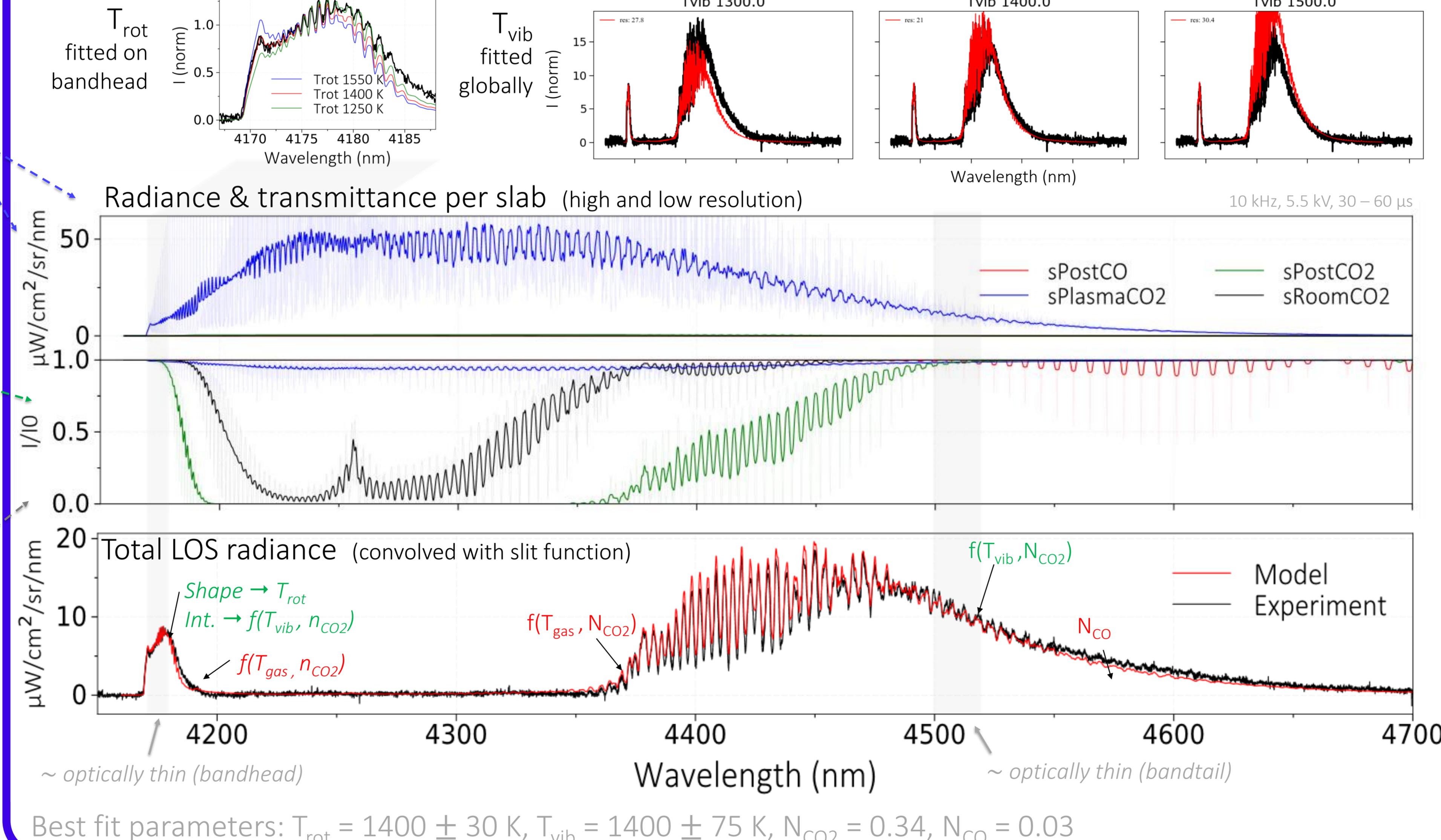
- Distribution: overpopulation factors allow arbitrary non equilibrium distributions of vibrational levels; rotational ~ Boltzman

LOS Model

Fixed & fitted parameters

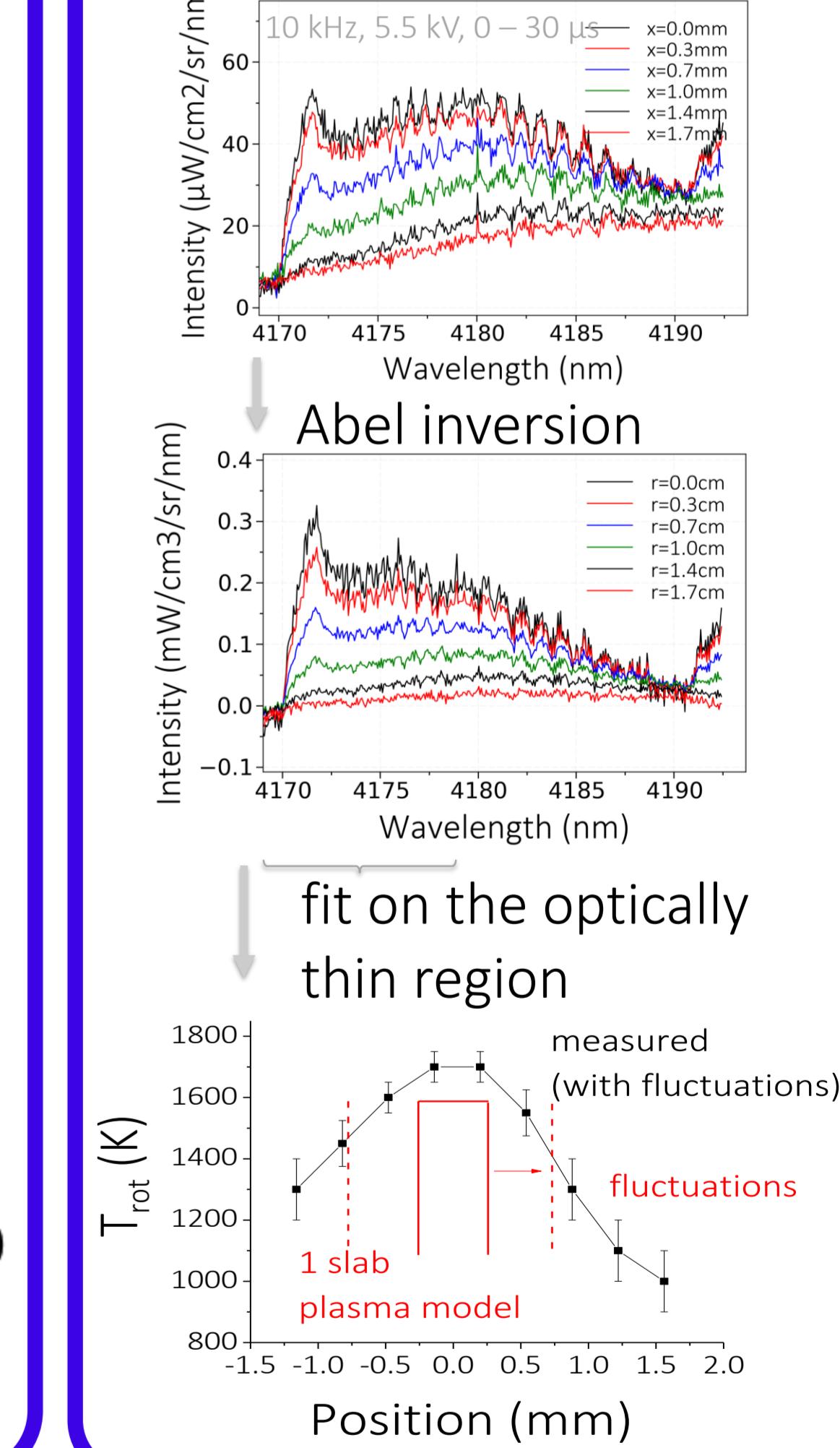


Fitting method



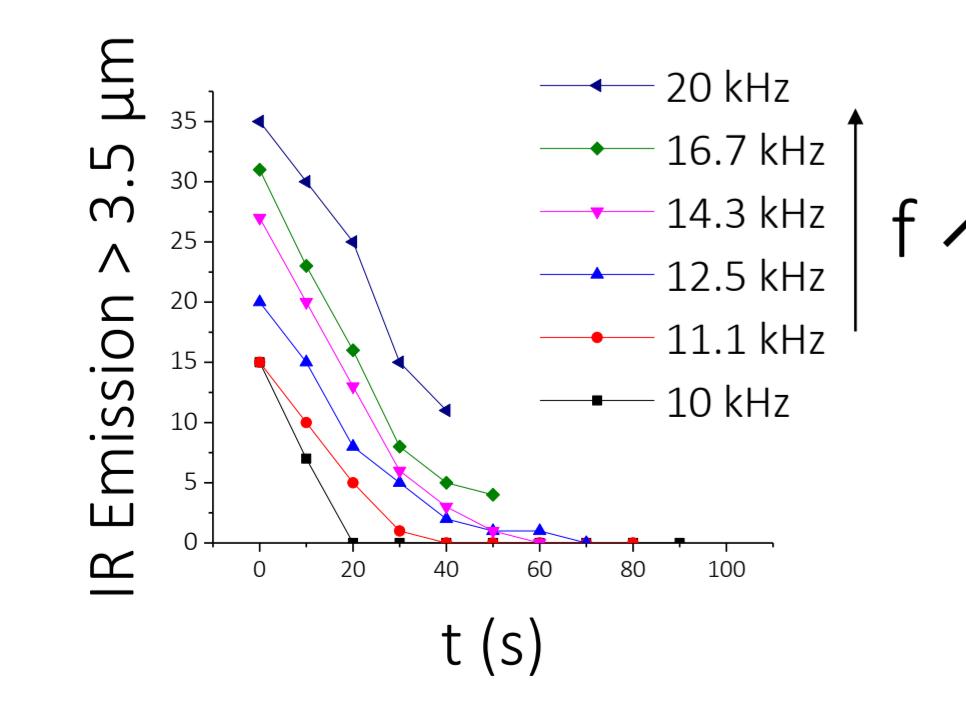
Refining

the LOS model with partial binning → 1D profile



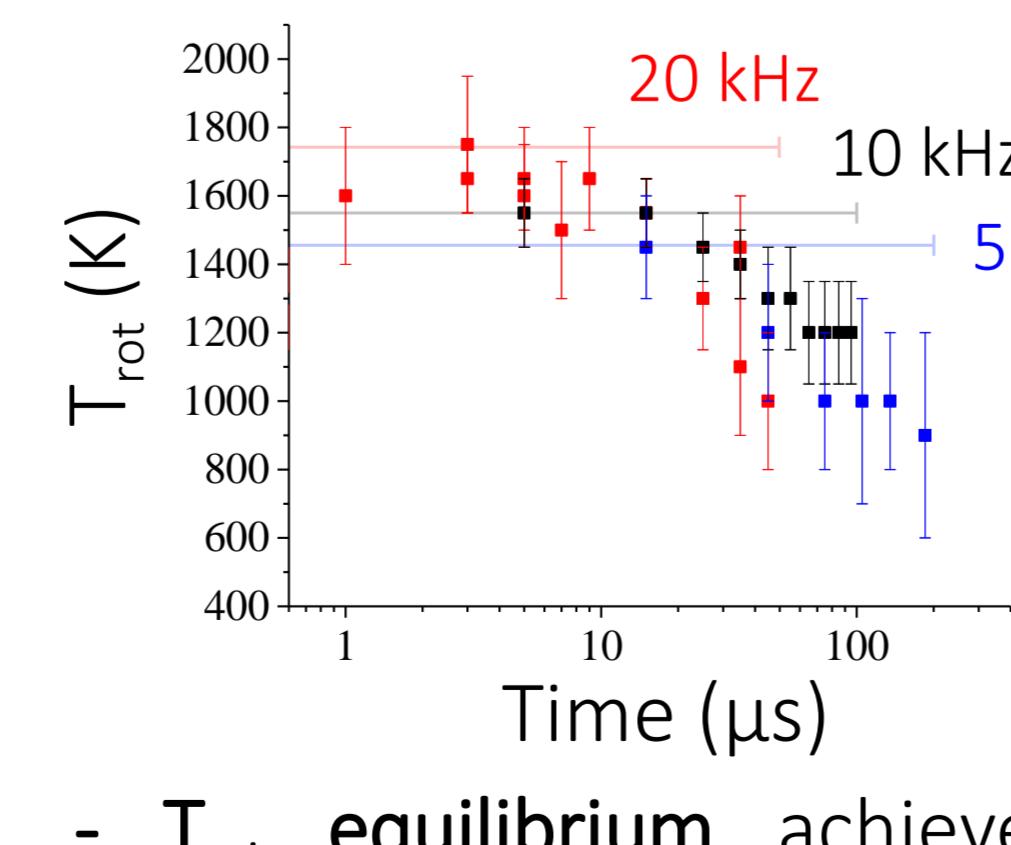
Results & Conclusions

Total IR emission (> 3.5 μm)



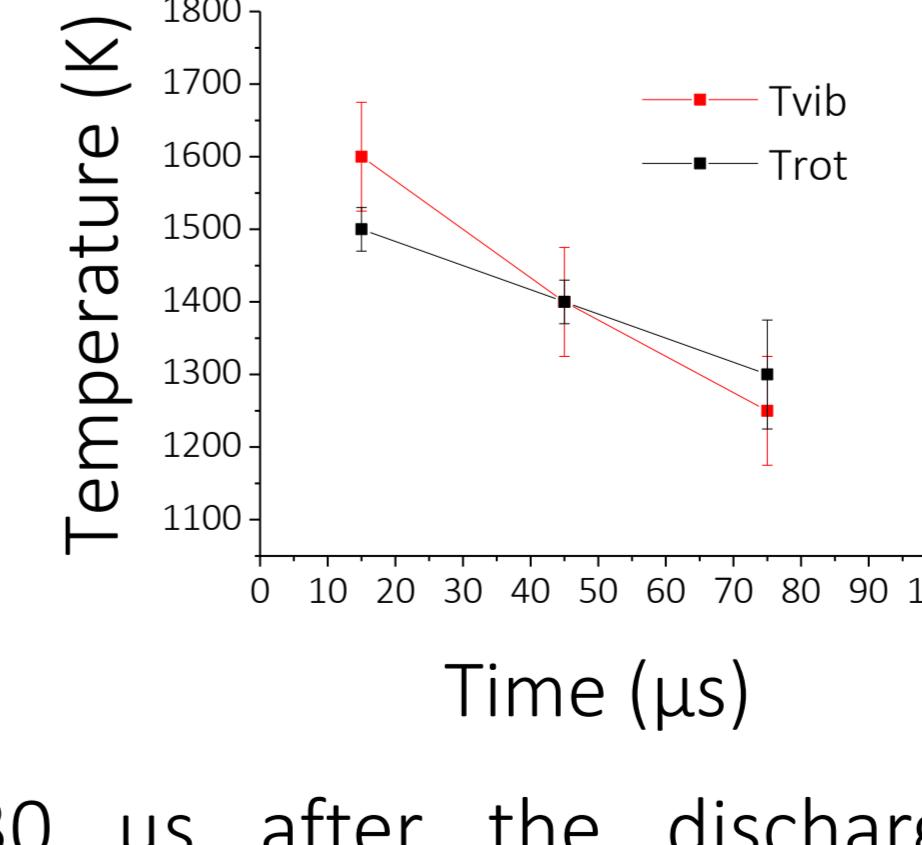
➤ Synergistic effect: vibrational excitation of CO₂ increases for frequencies $f > f_c \sim 12 \text{ kHz}$

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

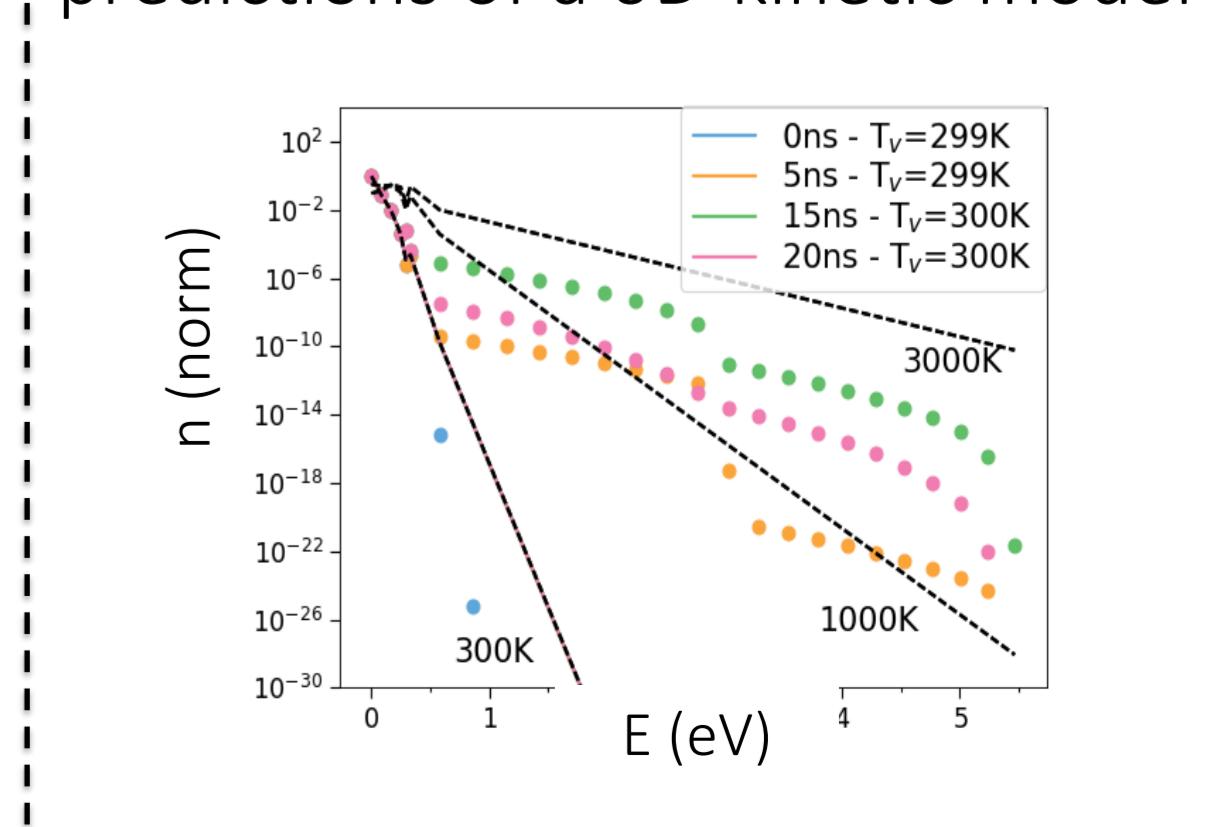


Fitting results:

- 10 kHz: $T_{\text{vib}} - T_{\text{rot}}$ equilibrium achieved within the first 30 μs after the discharge. Correct modelling can be obtained with a simplified 3-slabs model
- 20 kHz: evidence of non-equilibrium in the first 30 μs 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



NRP, vibrational specific OD model)