



Physikalisch-Technische Bundesanstalt
National Metrology Institute

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Infrared Intrapulse Spectroscopy

The first part of the poster covers the laser technology followed by two applications, one at PTB in Germany and the other one at UCLA in the USA

Laser technology

How the emission wavelength chirps during a pulse

At the beginning of the laser pulse, the current injected in the laser chip heats the laser. When the temperature increases, emission of the DFB laser remains single mode but the central frequency decreases in wavenumbers because of the temperature dependence of the refractive index. For this reason, the laser emission wavelength shifts with time inside the current pulses (see Figure 1 and the formula in Figure 2.)

The initial frequency at the beginning of the pulse can be chosen by setting the chip temperature at a certain value before pulsing the laser. Indeed, the operating temperature of a given chip can be varied within a certain temperature range, as illustrated in Figure 6.

As seen in Figure 3, the wavelength changes more slowly at the end of the pulse than at the beginning of the pulse, we see that the laser frequency decreases by 4.5 wavenumbers in the first microsecond but only by 0.5 wavenumbers in the second half of the 2μs-long pulse. This is because the laser temperature is stabilizing after a while and therefore, the wavelength doesn't shift as much. If the pulse were extremely long, we'd be operating the laser in CW and the emission wavelength would be stabilized.

The optimal pulse width providing wide wavelength tuning is of the order of a few μs provided the duty cycle is kept below ~20% .

As seen in Figure 3, the optical power remains nearly constant during the pulse under typical operating conditions.

Up to 25μm achievable in emission wavelength

In pulsed mode, the InAs based QCLs emission wavelength can reach 25μm (see Figure 4). This is due to the fact that the InAs-based quantum cascade lasers have a lower current threshold density compared with the more common quantum cascade lasers based on InP (see Figure 5).

Definition:

Intrapulse Chirp: The shift in emitted wavelength of the laser during the pulse. It can also represent the value, in wavenumbers, of the total wavelength shift during the pulse (also called **tunability**).

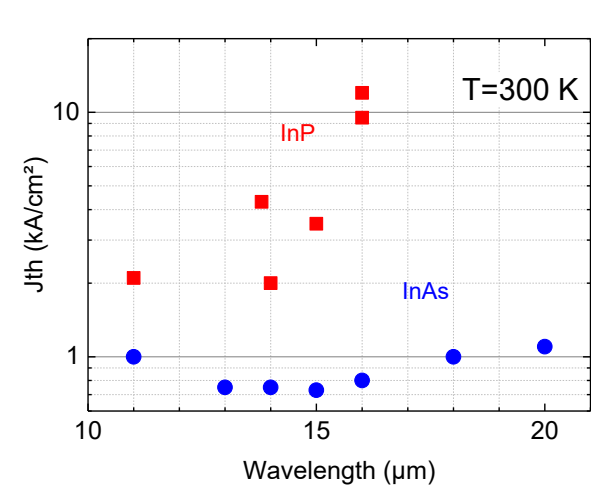


Figure 5. Threshold current density of InAs-based QCLs compared to InP-based QCLs for different wavelengths with no facet treatment. InAs/AISB QCLs outperform InP-based lasers at long wavelength.

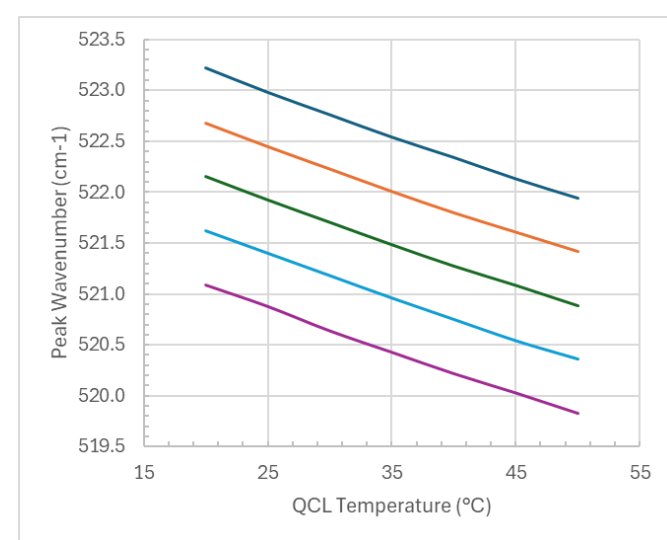


Figure 6. Spectral tuning of a DFB laser. Peak wavenumber of the laser as a function of the chip temperature.

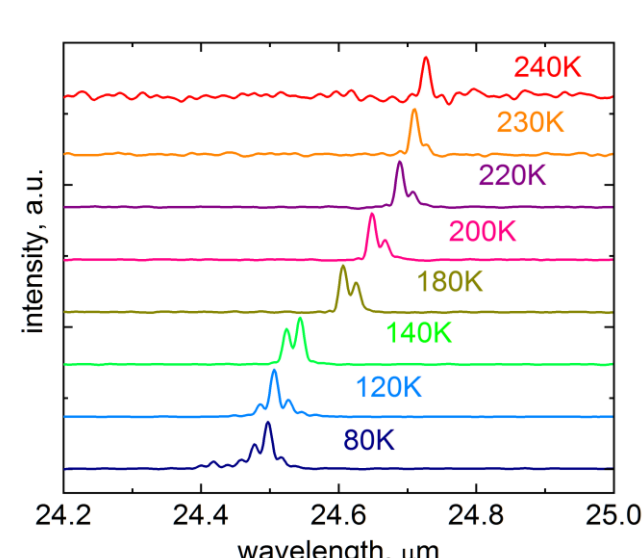


Figure 4. Emission spectra close to 25 μm wavelength at temperatures up to 240K.

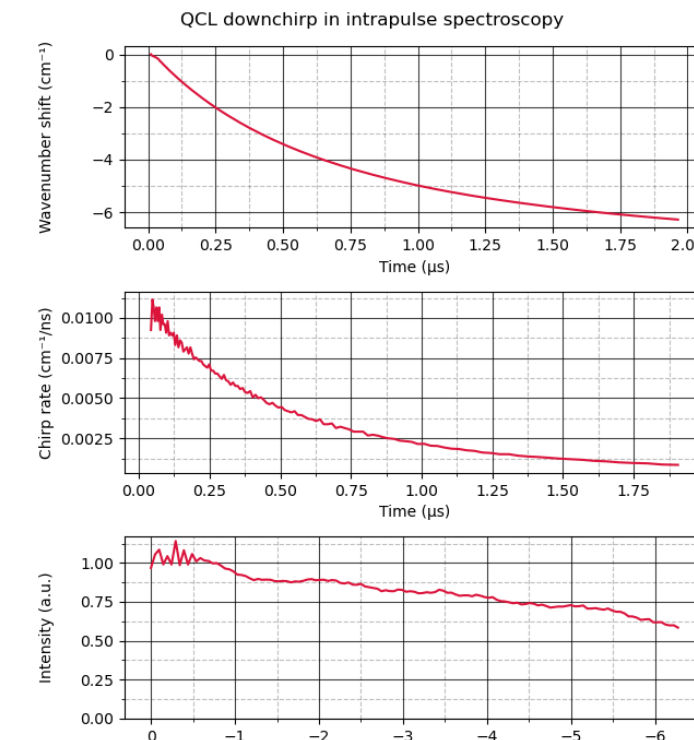


Figure 3. Intra-pulse chirp, wavelength chirp speed and optical power as a function of the chirp for a laser emitting at ~1285cm⁻¹. This is a typical behavior of lasers, even in the far infrared. The pulse is 2μs long with a pulse interval of 500μs.

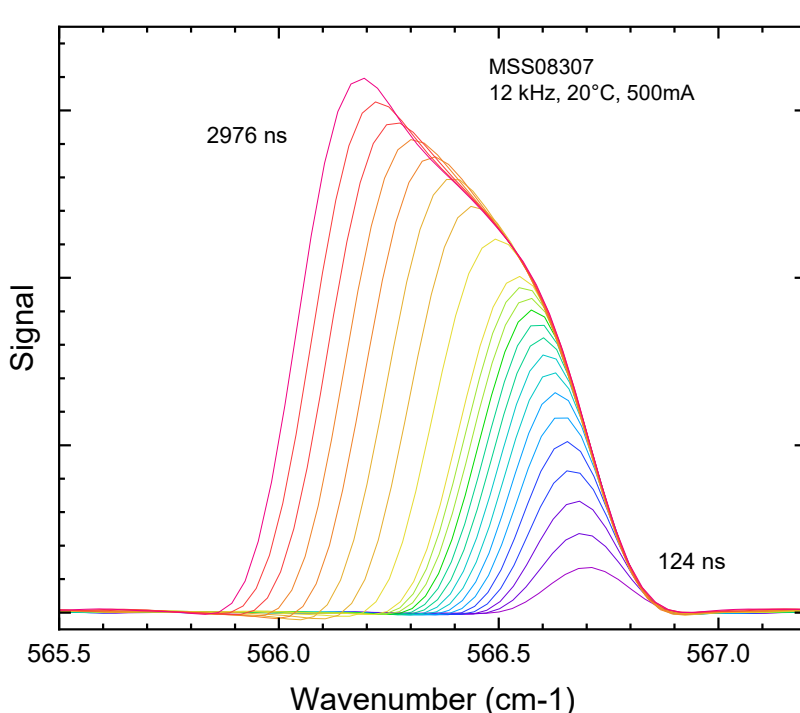


Figure 1. Intrapulse chirp of a ~17μm laser. Measured spectra with increasing pulse length. We see that the intra-pulse chirp is ~0.65cm⁻¹ for a 3μs pulse. The amplitude of the spectra of the longer pulses is higher than the amplitude of the spectra of the shorter pulses because during the longer pulses, more photons are emitted and therefore the recorded spectra is bigger. We see that as the pulse length increases, more photons are emitted at lower wavenumbers. This is because a laser that heats will emit photons will less energy, thus decreased wavenumbers.



Current density J , voltage V
Injected power $P = V \cdot J \cdot L \cdot w$
Active region volume $V_{ol} = L \cdot w \cdot h$
Volumetric heat capacity of the active region C_v

$$dT = \frac{P}{C_v \cdot V_{ol}} \cdot dt = \frac{V \cdot J}{C_v \cdot h} \cdot dt$$

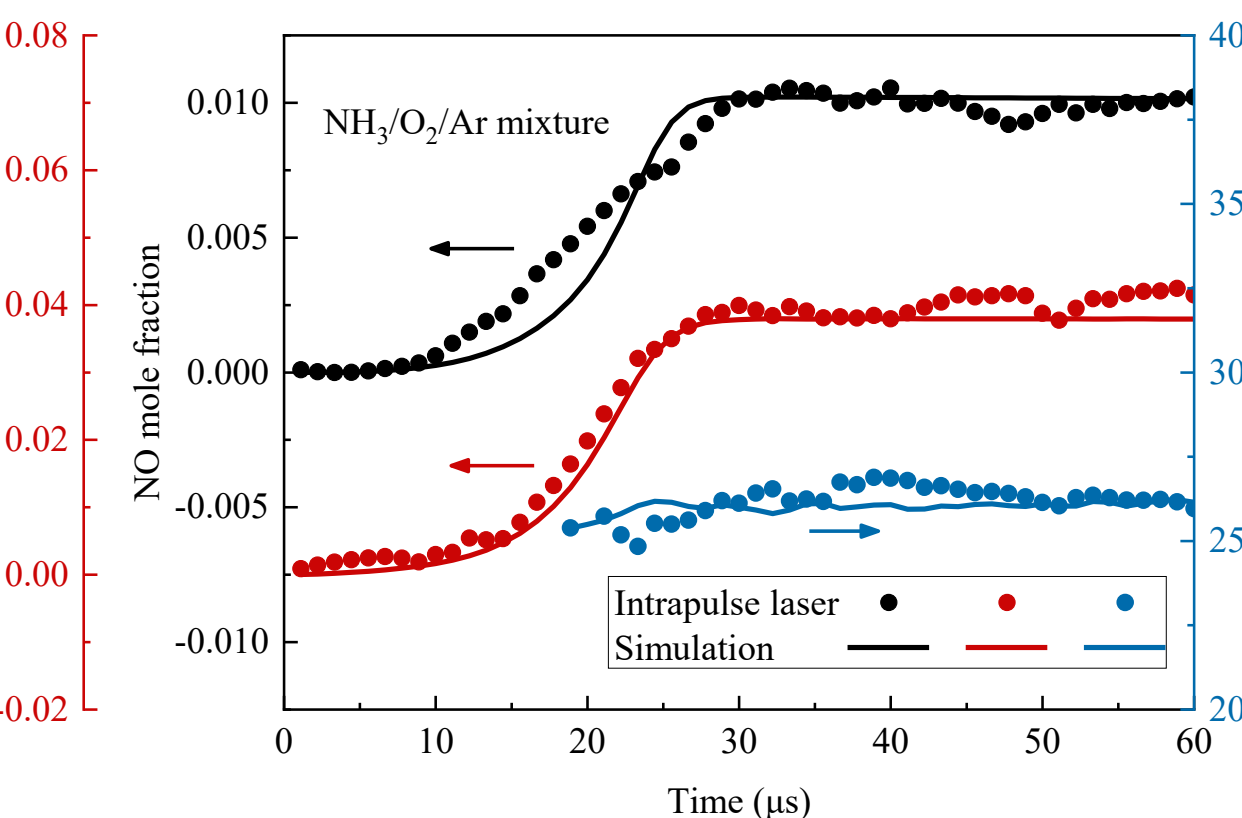
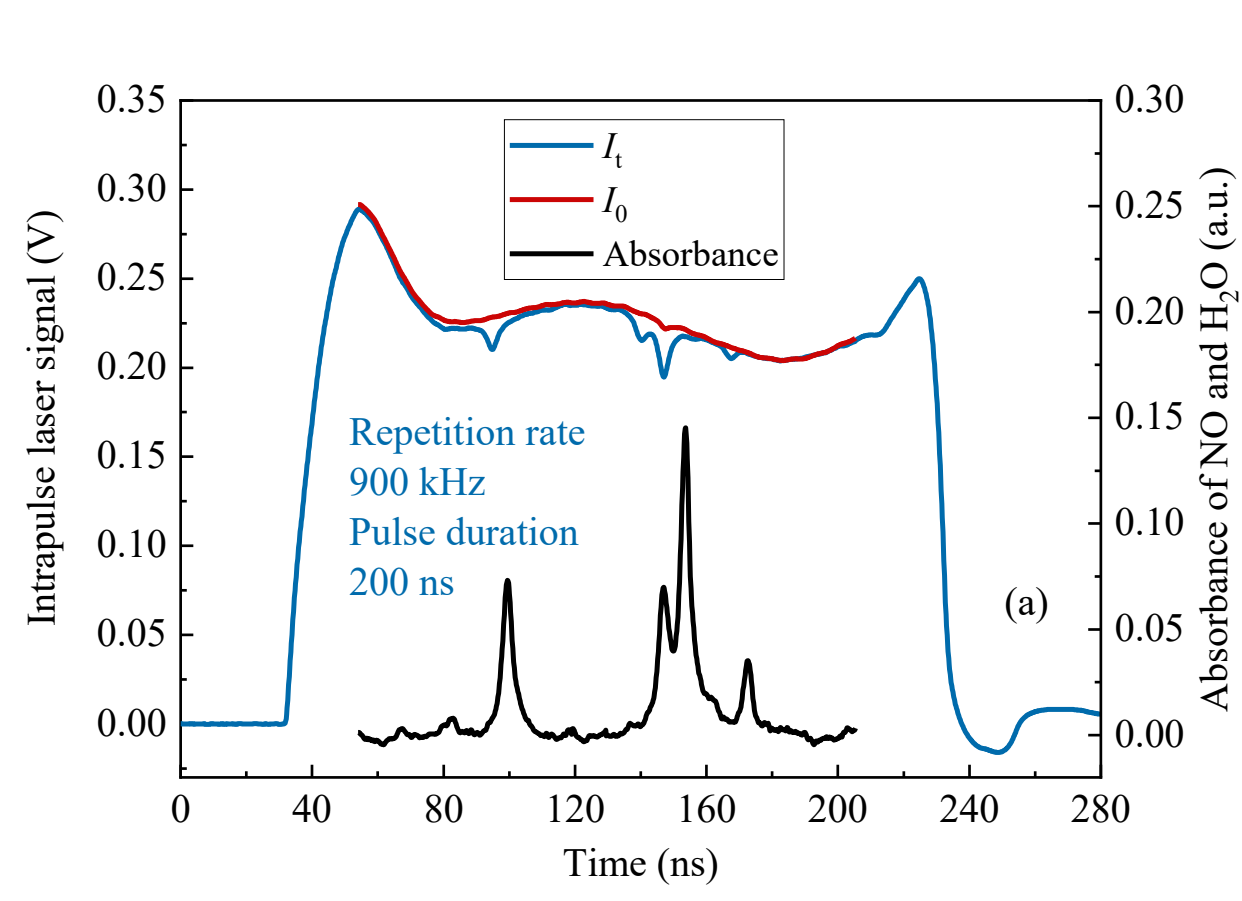
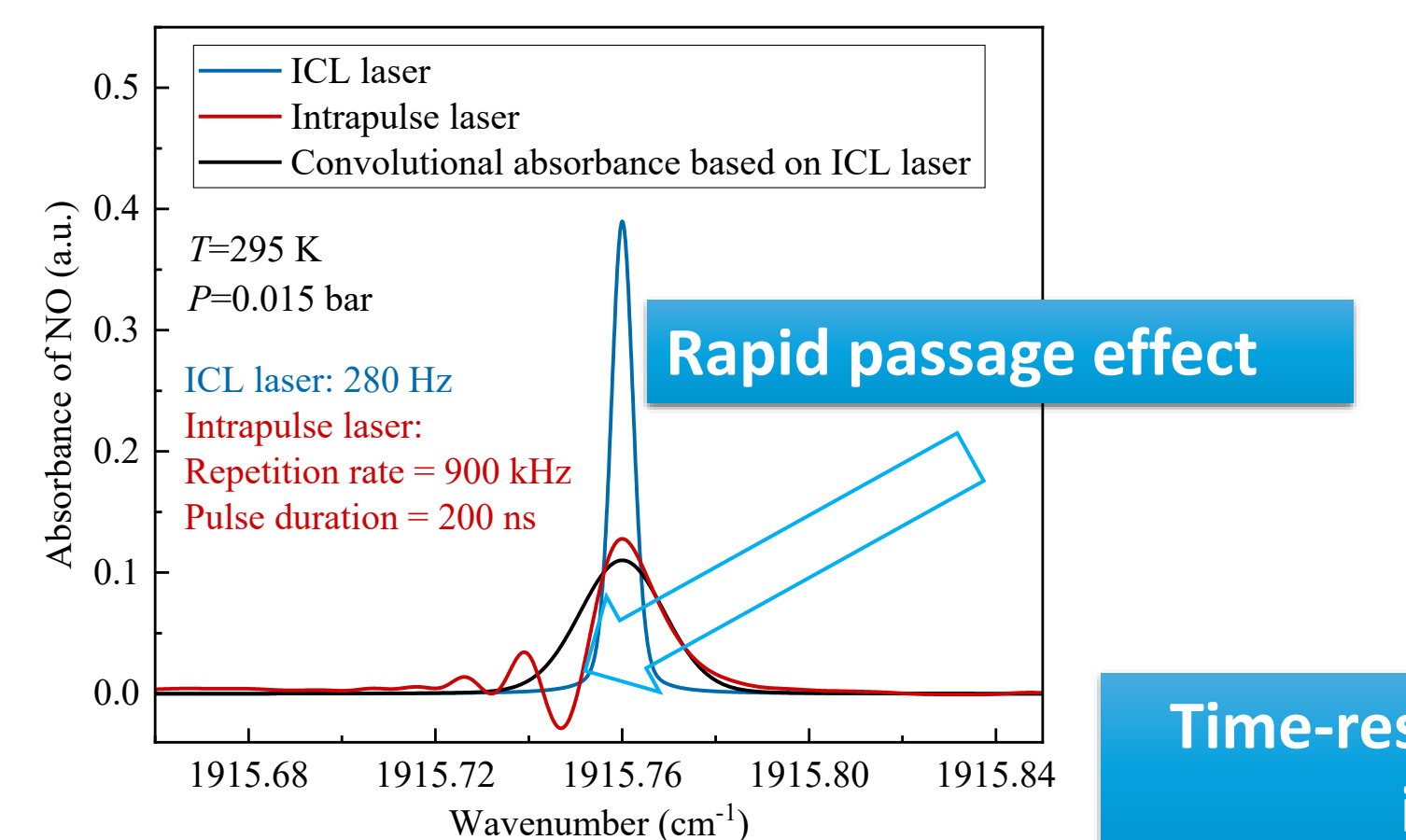
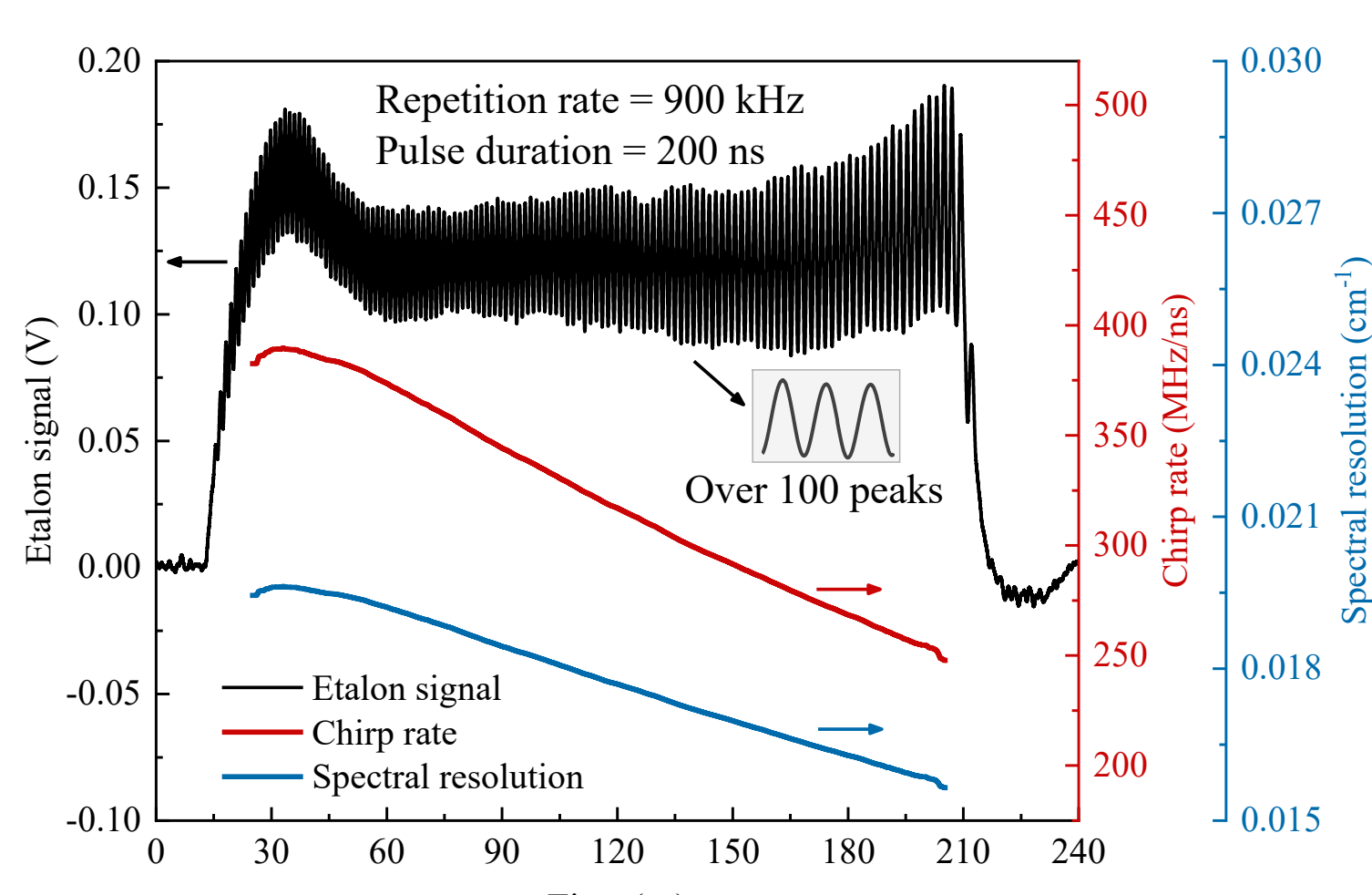
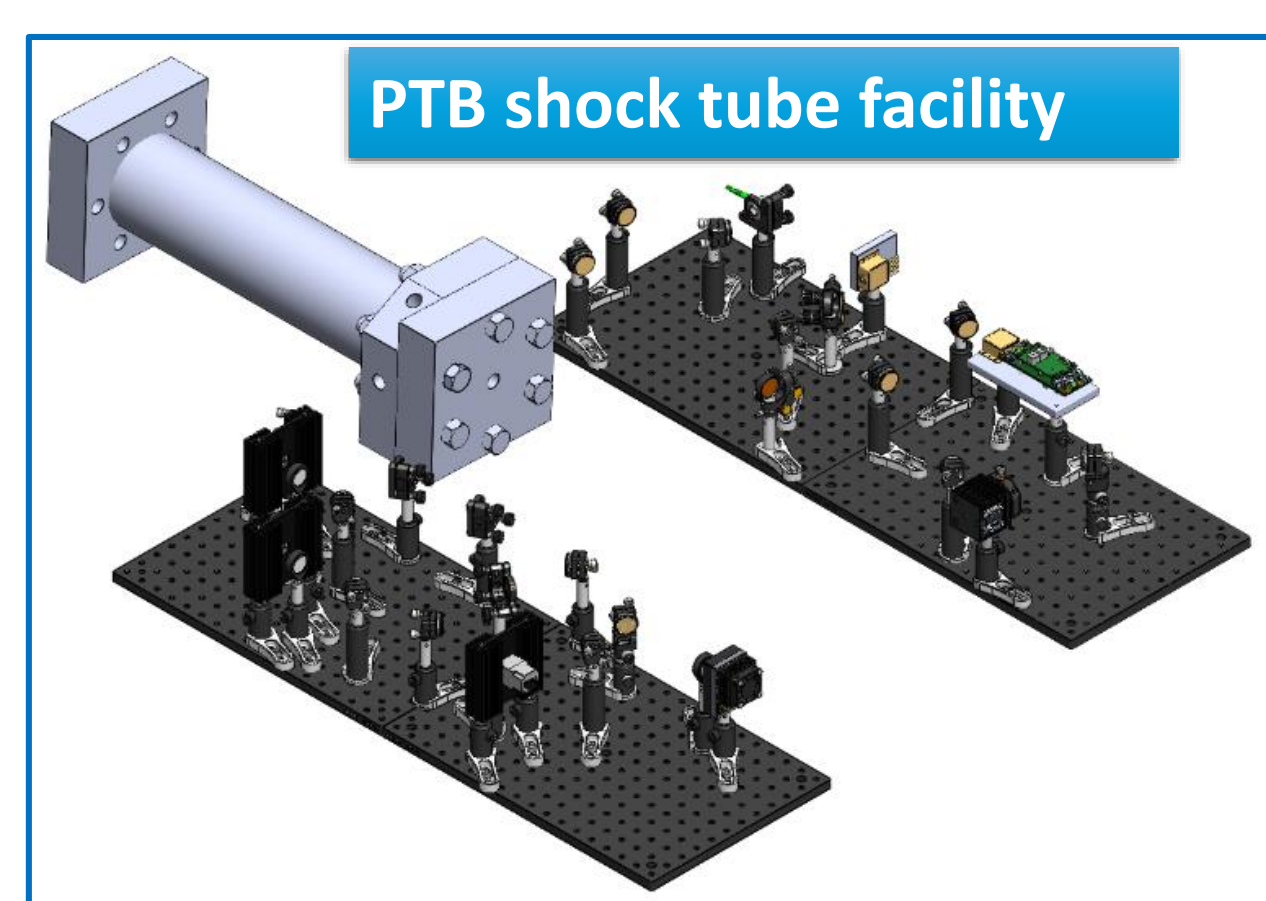
$$\frac{1}{\sigma} \frac{d\sigma}{dt} = -\frac{1}{n} \frac{dn}{dt}$$

$$d\sigma = -\frac{1}{n} \frac{dn}{dt} \cdot \frac{V \cdot J}{C_v \cdot h} \cdot dt \approx C_{te} \cdot \frac{1}{\lambda^2} \cdot V \cdot J \cdot dt$$

$\sim C_{constant}$ $\sim 1/\lambda$ $\sim \lambda$

Figure 2. Formula for the intra-pulse chirp $d\sigma$. The intra-pulse chirp scales with $1/\lambda^2$ so this means that the **chirp is smaller at longer wavelengths**. For example, the chirp at 15μm wavelength is <1/10 the chirp at 5μm wavelength.

Simultaneous dynamic NO, H₂O and temperature measurement with MHz time resolution



Time-resolved NO, H₂O and temperature measured by intrapulse laser and simulation results



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Absorbance of NO and H₂O spectra during NH₃ oxidation in the shock tube

- Validated intrapulse laser spectroscopy for NO, H₂O, and temperature.
- Measured spectrum with MHz resolution with 250-400 MHz/ns chirp rate.
- Enabled fast, calibration-free multi-parameter sensing.
- Excellent agreement with ICL NO and DFB H₂O laser diagnostics

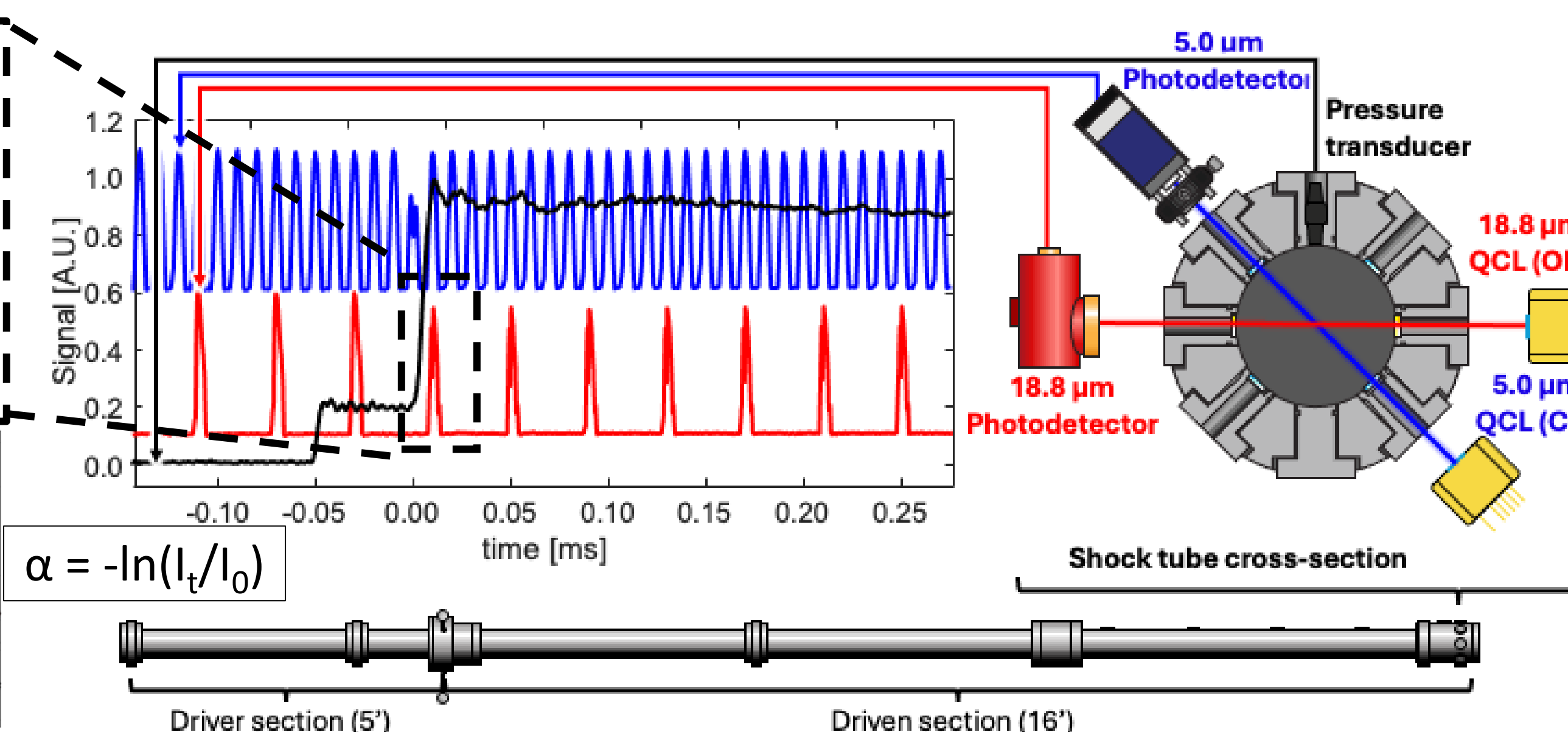
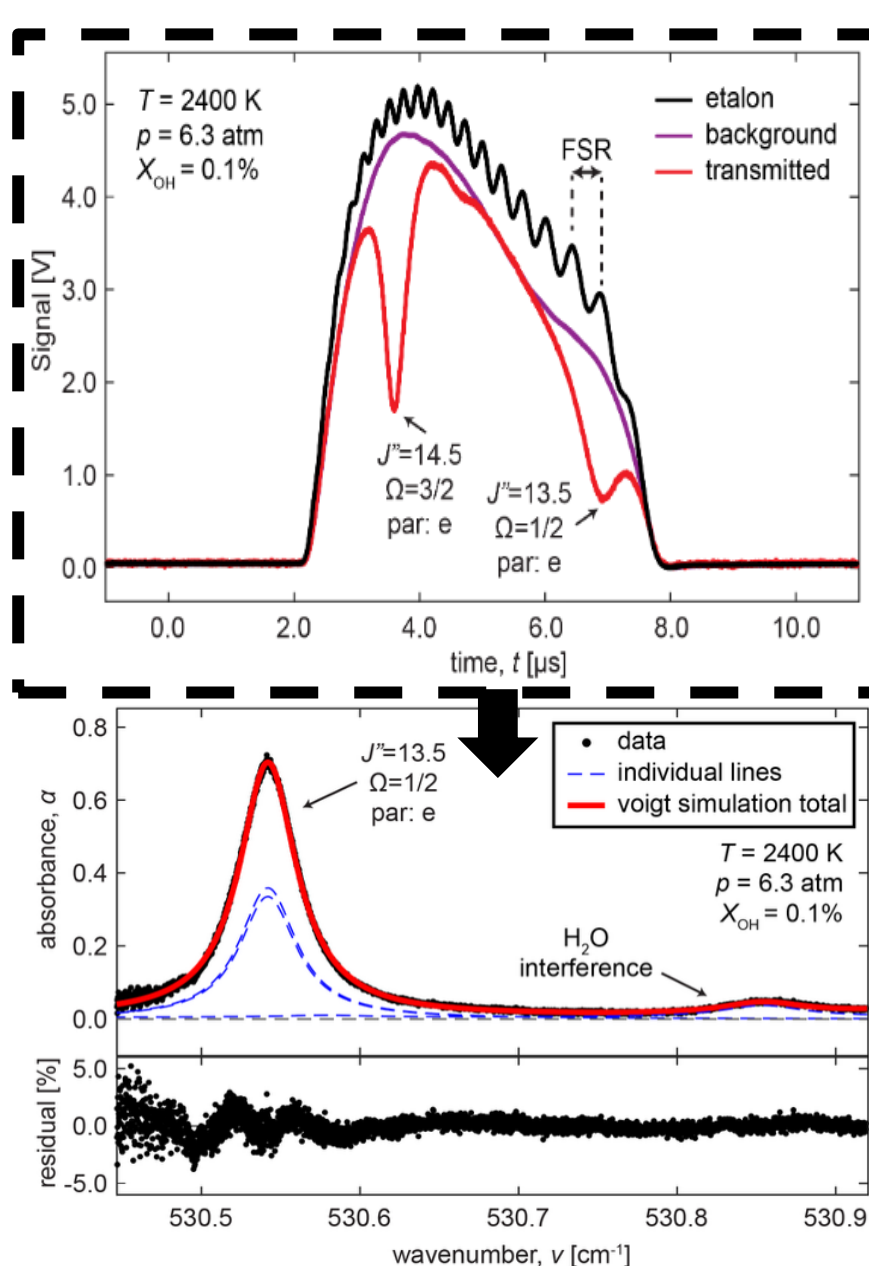
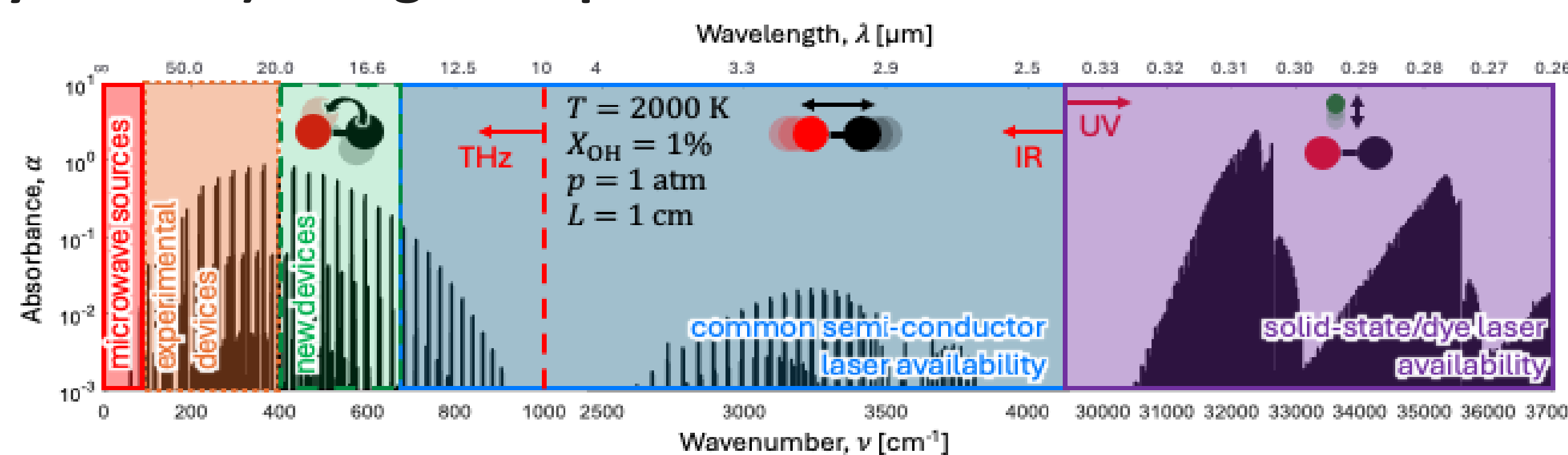
SCAN ME



The content included in this section is derived from published work: Zhu, D., Seifert, L., Agarwal, S., Shu, B., Fernandes, R. and Qu, Z., *Sensors and Actuators B: Chemical* (2025): 138596.

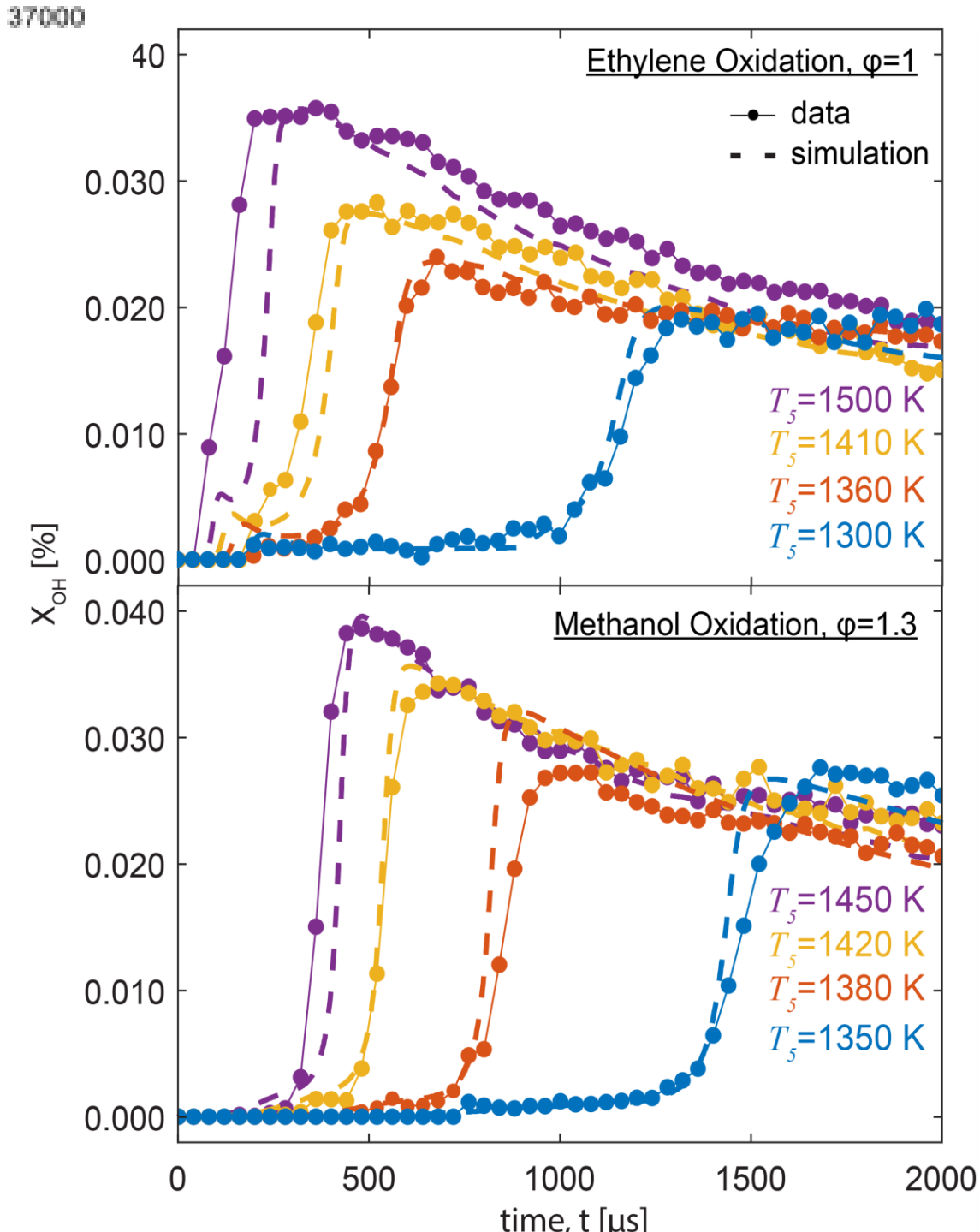
THz rotational absorption spectroscopy of OH (hydroxyl radical) at high temperatures

- The hydroxyl radical (OH) is a key intermediate in combustion
- OH THz spectra ~50x stronger than MIR spectra
- Pure rotational OH lines accessible with rapidly-tuning semiconductor source at elevated temperatures
 - Enables spectrally-resolved measurements
 - Advantageous to fixed-wavelength / slow-tuning methods (typical of UV sources)



Chemical Kinetics Species Measurements

- Measured reference line strengths enable quantitative OH mole fraction measurements
- Time resolution of 25kHz associated with pulse rate
- 5 ppm detection limit at 10 cm optical pathlength
- Good agreement with independent chemical model



The content included in this section is derived from published work: Kuennig N.M., Minesi N.Q., Honaker B.A., Spearrin R.M., *PROCI* (2024): 105480.



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