

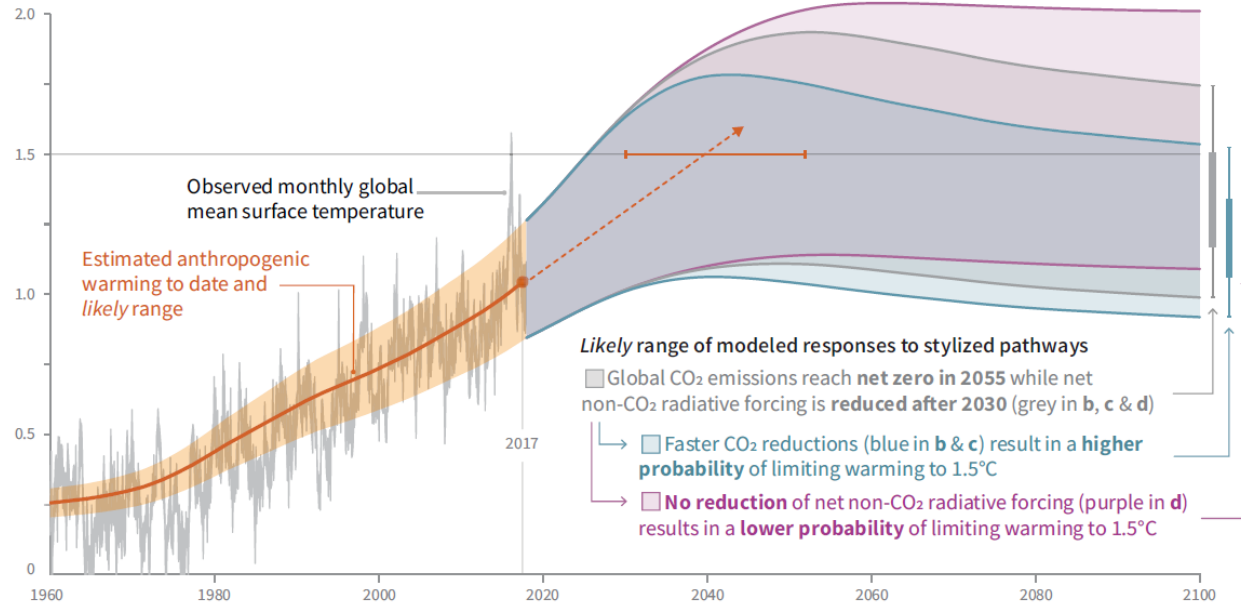
Mid-infrared tunable diode laser spectroscopy for the detection of fugitive methane emissions

Jalal Norooz Oliaee, Nicaulas Sabourin, James A. Gupta, Prem
Lobo, Kevin Thomson, Greg Smallwood

Council for Optical Radiation Measurements 2019
Ottawa, Canada

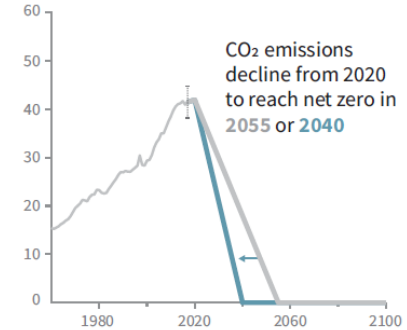
Motivation: Climate Impacts

Global warming relative to 1850-1900 (°C)

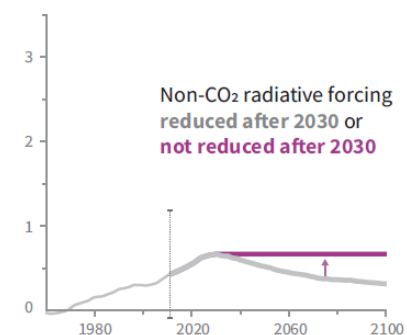


IPCC 2018, Special Report: Global Warming of 1.5 °C

Billion tonnes CO₂ per year (GtCO₂/yr)



Watts per square metre (W/m²)



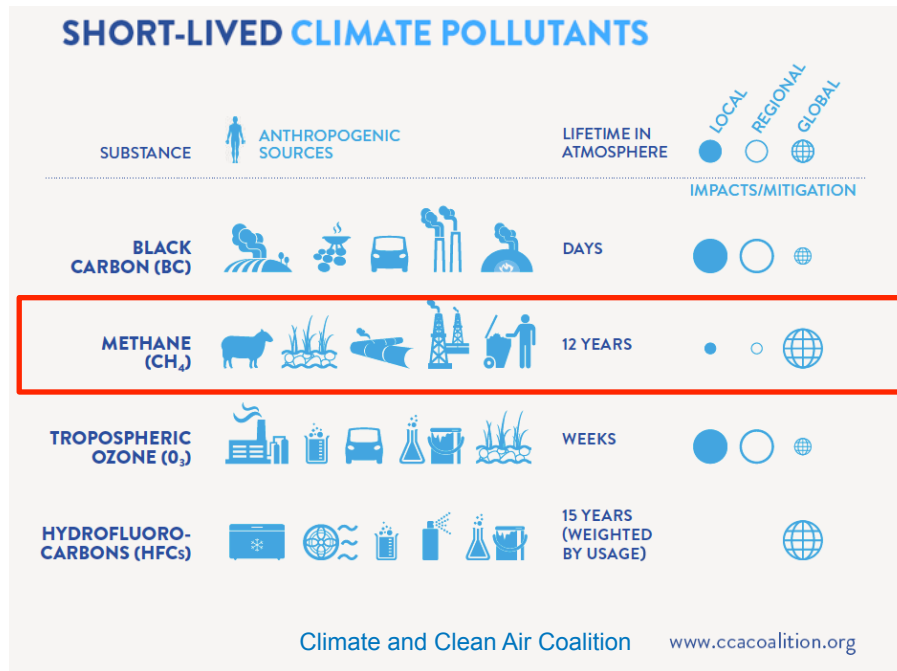
Non-CO₂ climate forcers

Short-lived climate forcing agents

- Black Carbon (BC)
- Methane (CH₄)
- Some fluorinated gases (e.g. HFCs)
- Ground-level ozone (O₃)

Long-lived climate forcing agents

- Nitrous oxide (N₂O)
- Some fluorinated gases (e.g. CF₄)



20/100-year
84/28 x CO₂

Global action on CH₄

IPCC 1.5°C Special Report, Section C.1.2.:

“Modelled pathways that limit global warming to 1.5°C with **no or limited overshoot** involve deep reductions in emissions of **methane** and black carbon (35% or more of both by 2050 relative to 2010).”

“Non-CO₂ emissions can be reduced as a result of broad mitigation measures in the energy sector.”

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“Non-CO₂ emissions can be reduced as a result of broad mitigation measures in the energy sector.”

Other motivations to reduce emissions:

For example:

\$\$\$ Lost methane is also lost potential energy and revenue \$\$\$

METHANE

A potent greenhouse gas with a short lifetime

Uncertainties in inventory estimates exist*

Gaps attributed to **unreported venting** or **fugitive** emissions due to episodic emissions, abnormal operating conditions, leaks, etc.

These emissions are often ignored but need to be **monitored**.

* M. R. Johnson et al. *Environ. Sci. Technol.* 51, 13008-13017 (2017)
R. A. Alvarez et al. *Science* 361, 186-188 (2018)

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UAV-based measurements

- Autonomous and flexible flight patterns
- Lower operation cost vs. manned aircrafts
- Near-surface operation
- Relaxed regulatory requirements

Requires sensitive, small, lightweight, power-efficient, and fast response time sensors.

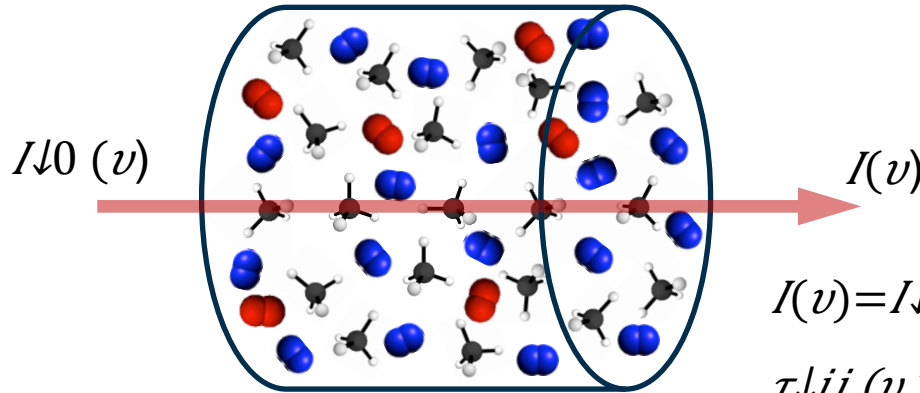


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SPECTROSCOPY

Wavelength Modulation Spectroscopy (WMS)
using
mid-infrared Tunable Diode Laser Absorption Spectroscopy (TDLAS)

Direct Absorption Spectroscopy (DAS)



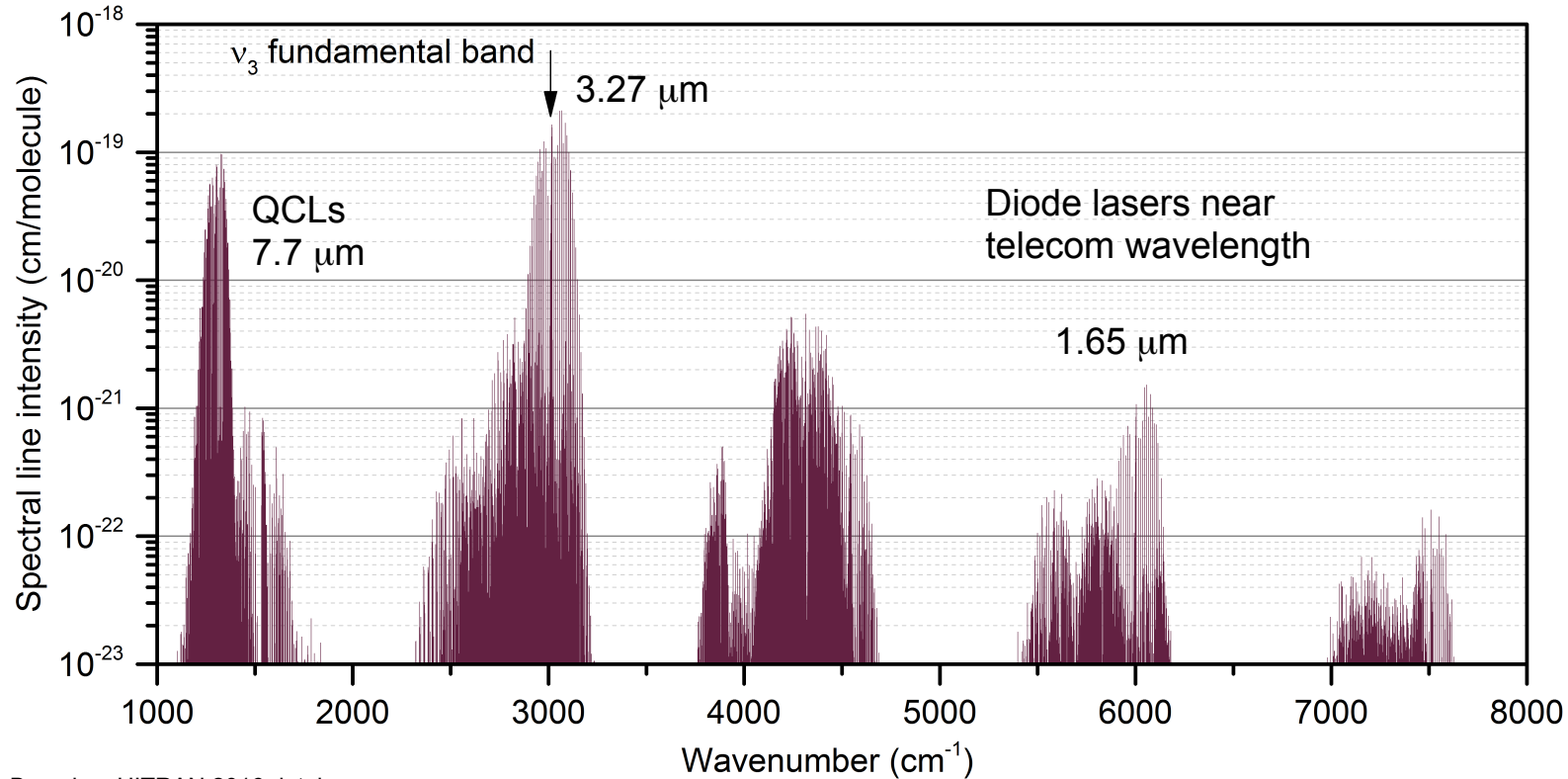
$$I(v) = I_0(v) e^{-\tau_{ij}(v, T, p)}$$

$$\tau_{ij}(v, T, p) = [CH_4] l S_{ij}(T) f(v; \nu_{ij}, T, p)$$

For fugitive emissions detection

- Strongest absorption line, S_{ij}
- Increase the absorption pathlength, l
- Wavelength modulation spectroscopy, $\nu = \nu_{ij} + a_M \sin(\omega_M t)$

Absorption bands of Methane



Based on HITRAN 2016 database

Mid-infrared light sources

Past

Mid-IR lead-salt diode lasers

- Multi-mode with mode-hops (i.e. dispersive optics)
- Cryogenic cooling (typically <130K)

Not suitable for portable applications

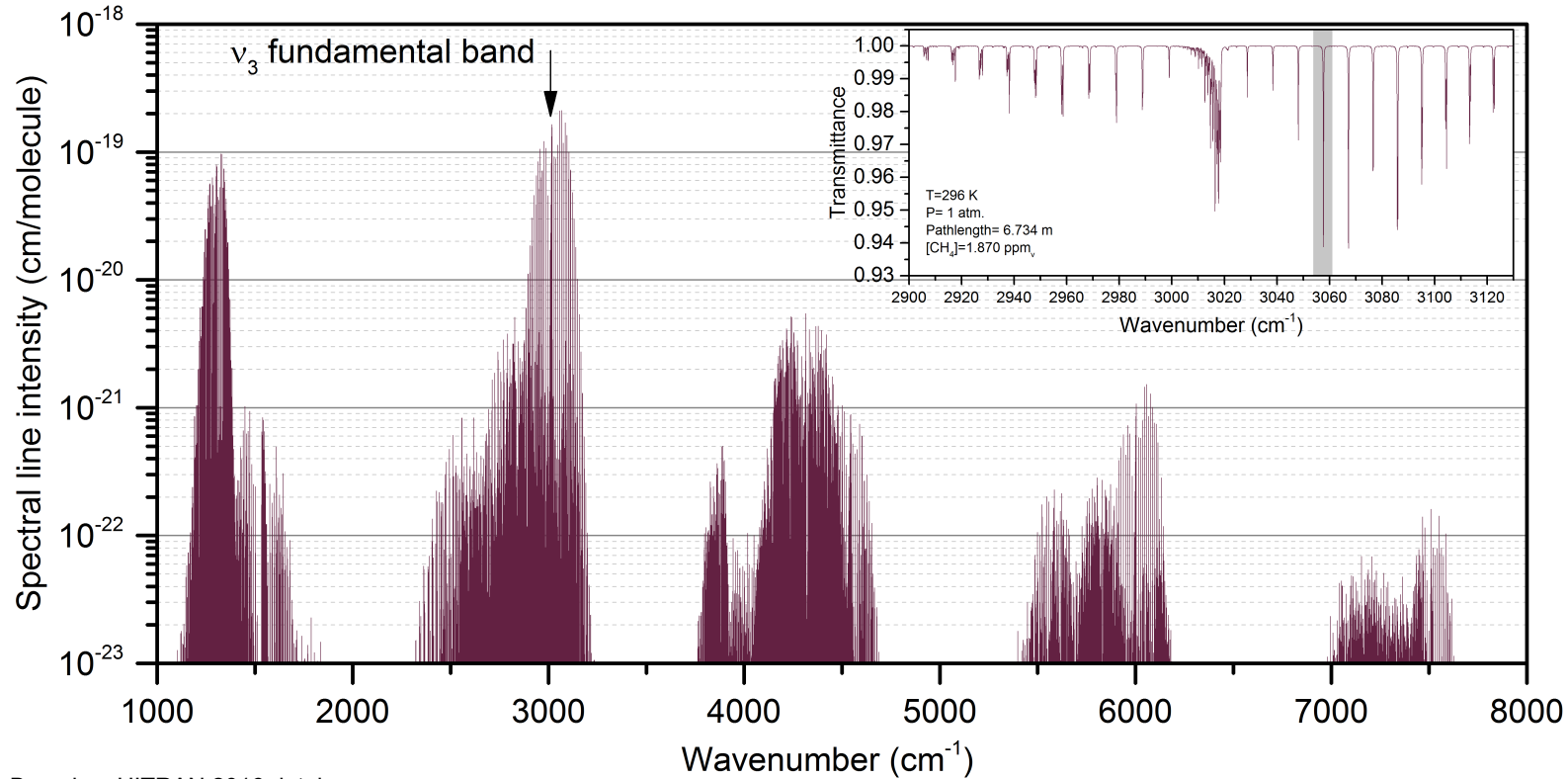
State of the art

GaSb-based DFB diode laser

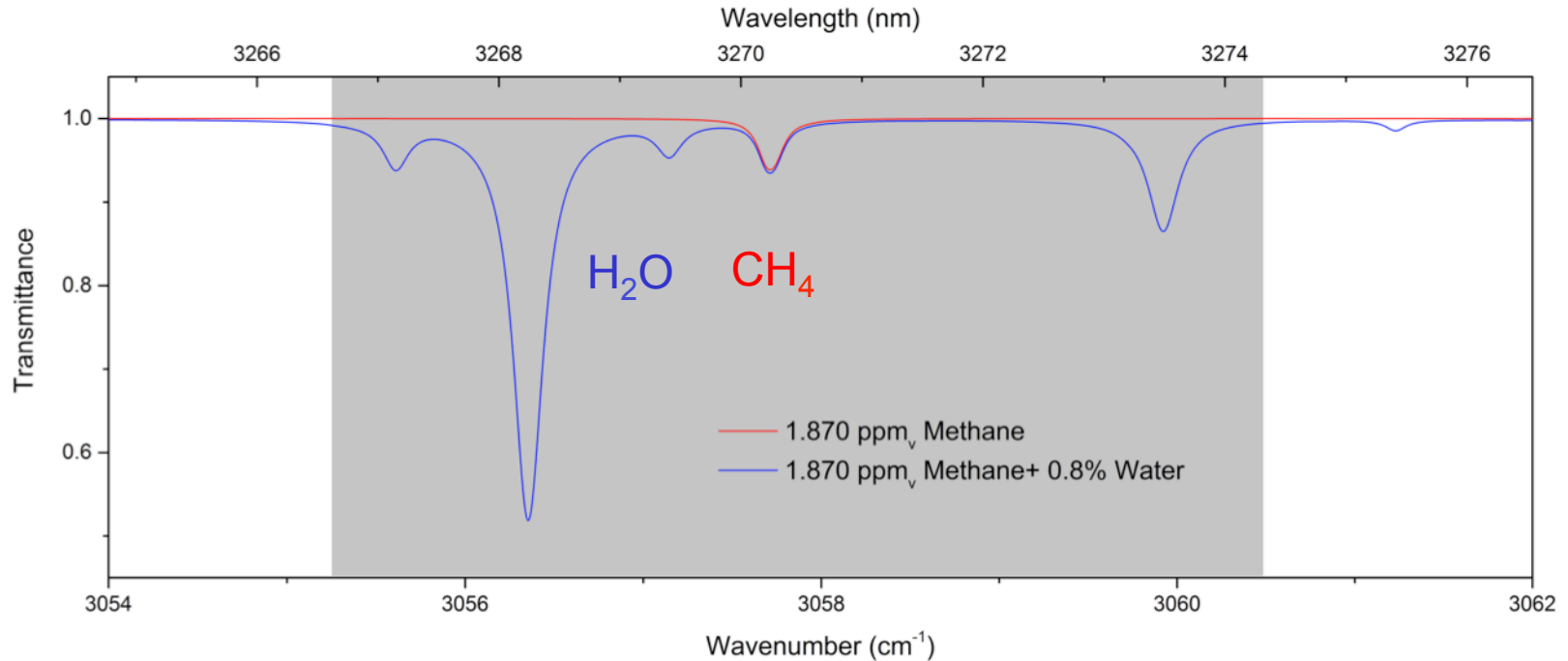
- Single mode emission: 3270 nm
- Near room temperature operation (TEC accessible)
- Small footprint
- Hermetically sealed

Suitable for space and mobile applications

Absorption bands of Methane



Spectral probing range

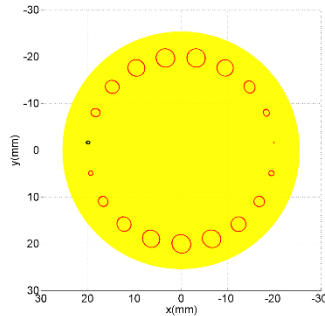
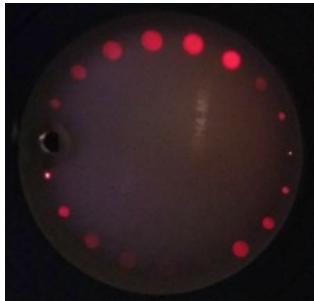


Calculated based on HITRAN 2016 database, P=1 atm., T=296 K, PL=6.734 m.

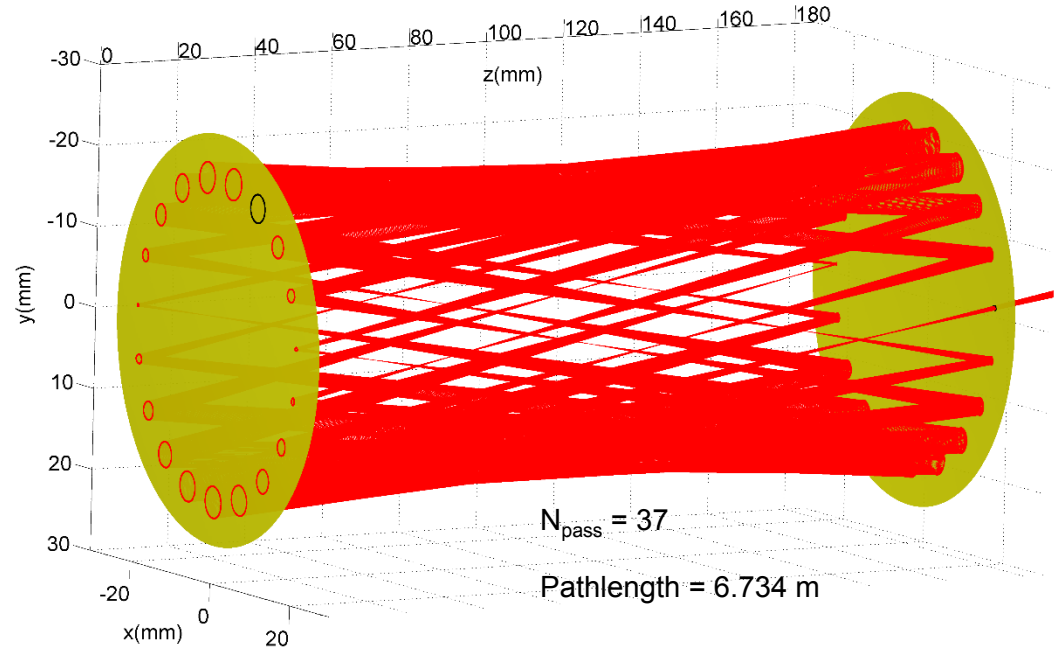
Multi-pass cell design

Herriott-type multi-pass cell

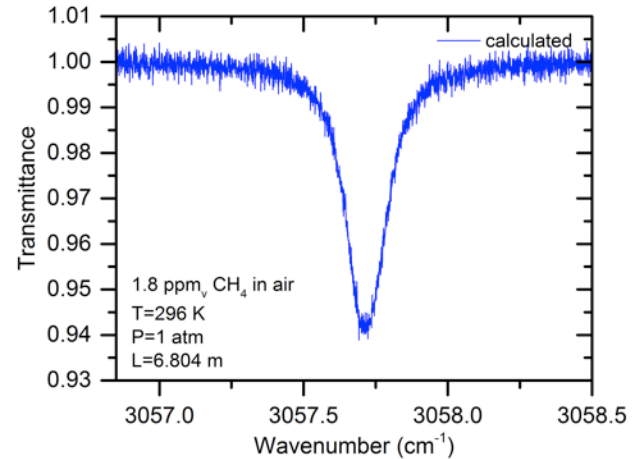
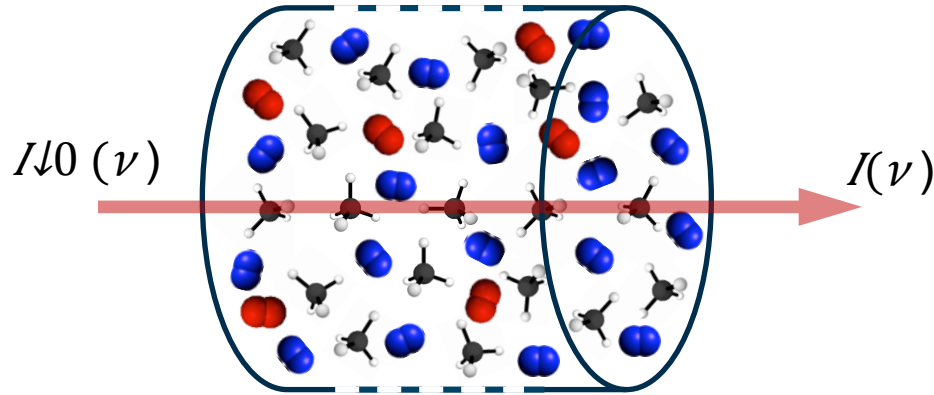
- 2-inch gold-coated ($R > 98.5\%$) spherical mirrors
- 37 passes, 6.734 meters of pathlength
- Open-path
- Not invoking the paraxial ray approximation



Spot pattern as seen from the back of the far mirror.



Direct Absorption Spectroscopy (DAS)



Optical depth about 0.06

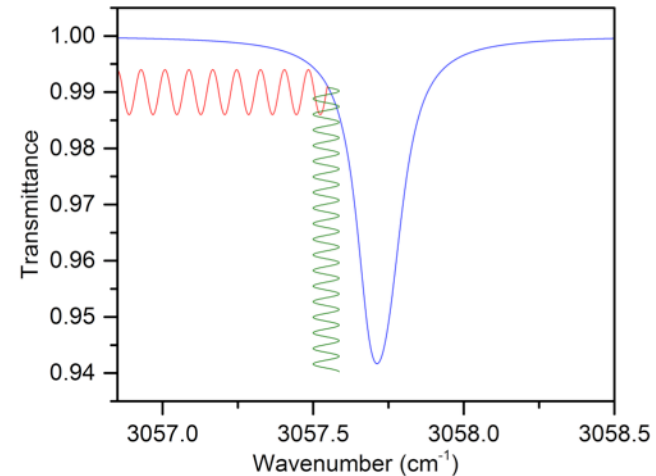
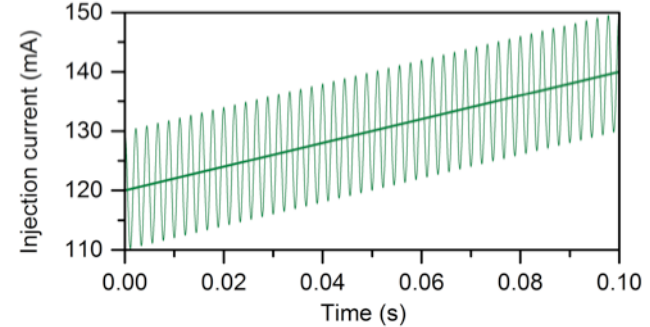
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Wavelength Modulation Spectroscopy (WMS)

Wavelength Modulation by TDL injection current

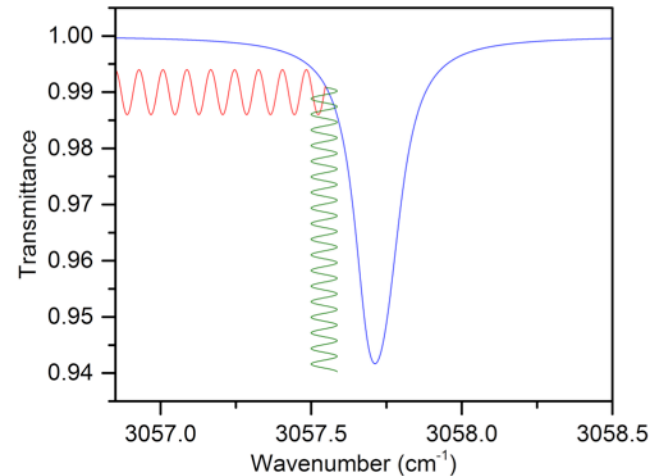
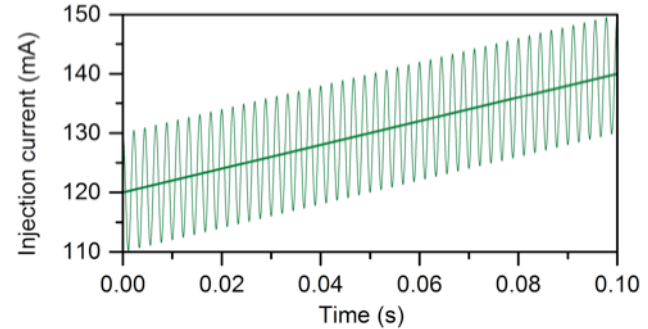
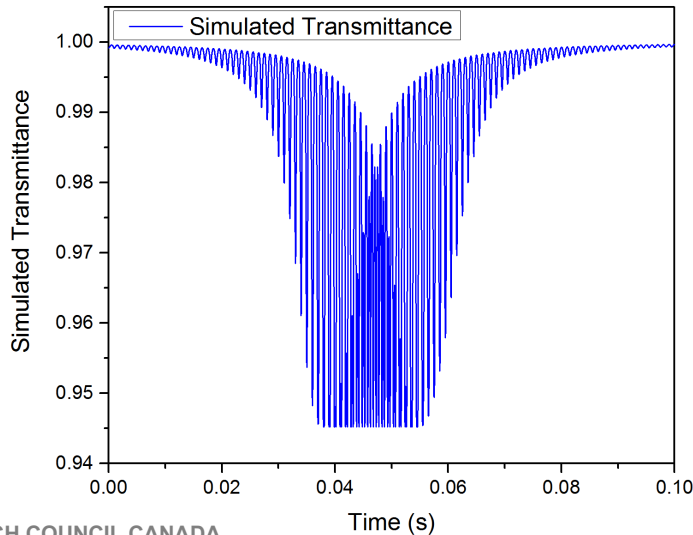
- Ramp for sweeping laser frequency
- High-frequency sine wave for modulation



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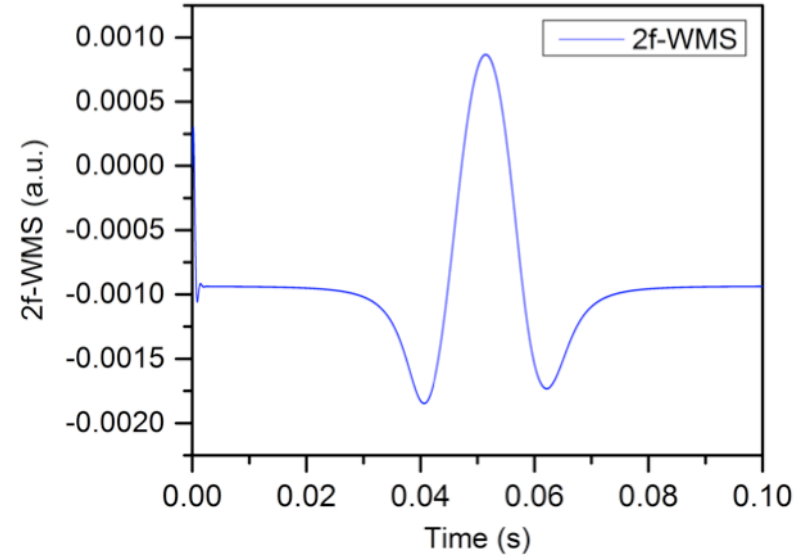
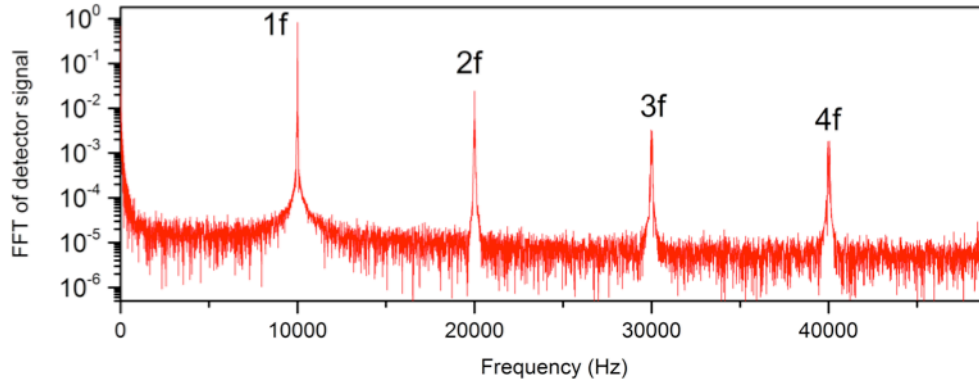
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Wavelength Modulation Spectroscopy (WMS)

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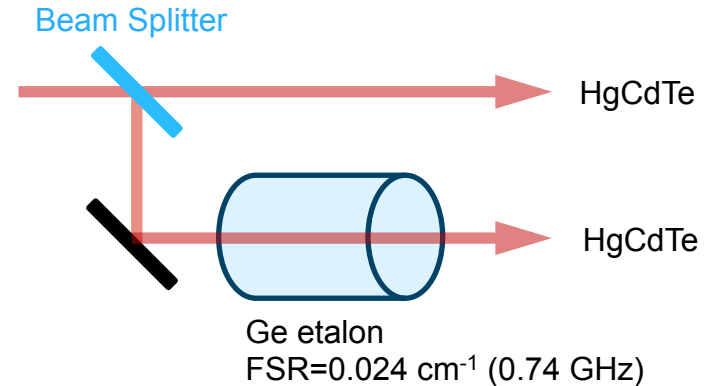
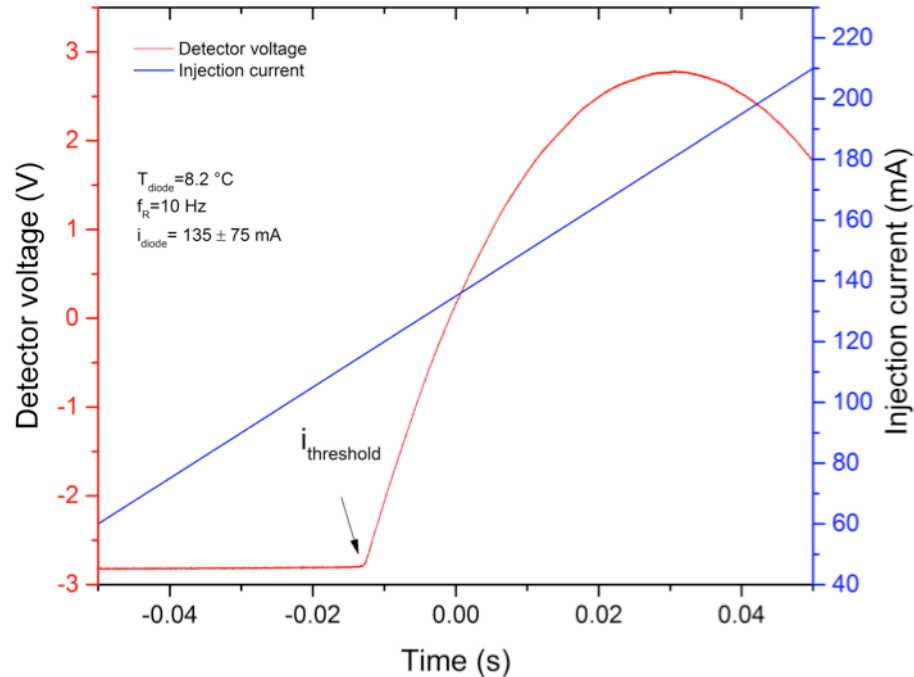
- Ramp for sweeping laser frequency
- High-frequency sine wave for modulation
- Detection at harmonics of the modulation frequency (i.e. lock-in amplifier)



$$S_{2f}^X(t) = LP\{I_{\text{det.}}(t) \times \cos(2\omega_M t + \theta)\}$$

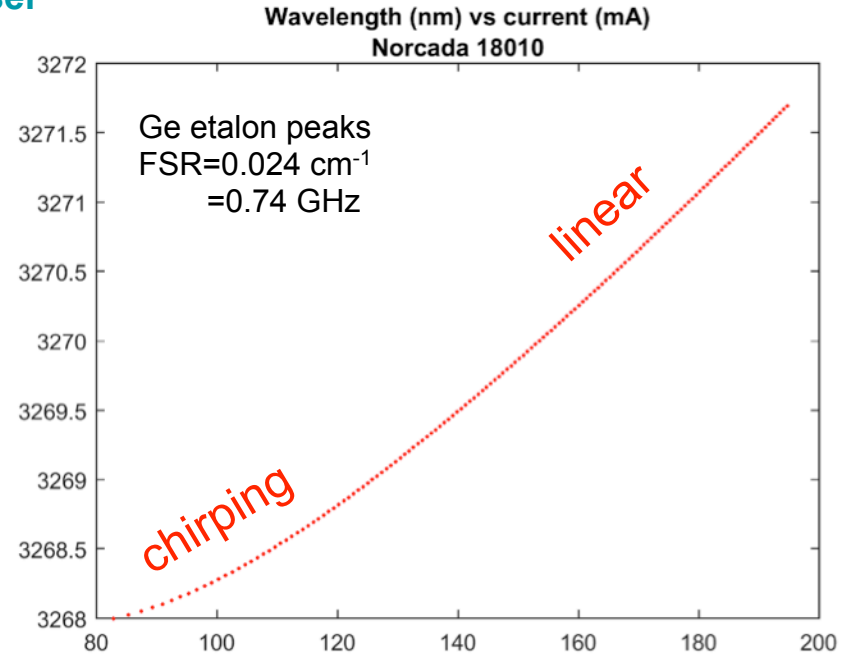
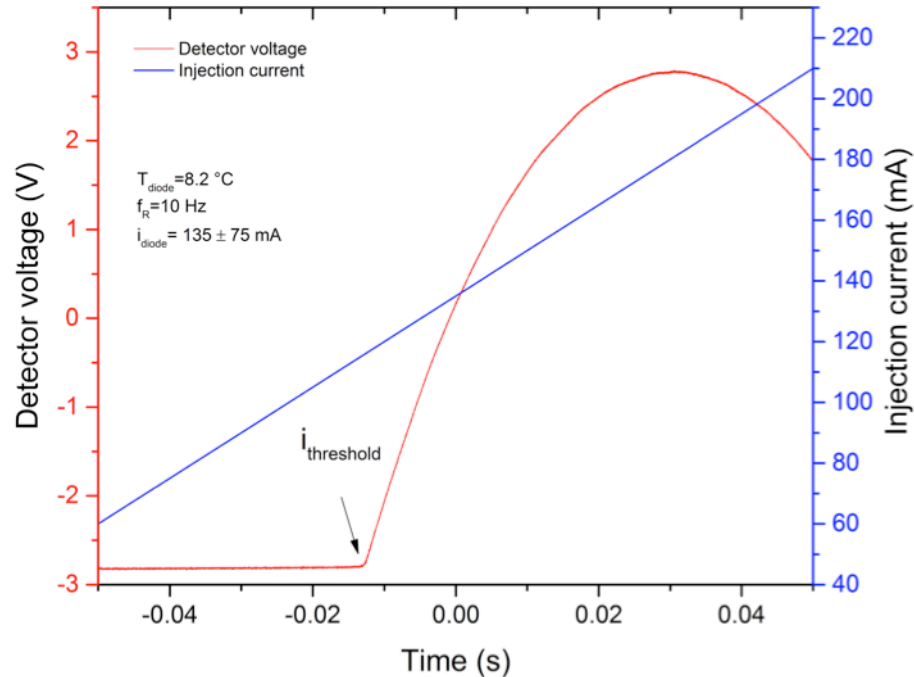
Characterization of the diode laser for WMS

Wavelength tuning characteristics of GaSb DFB laser



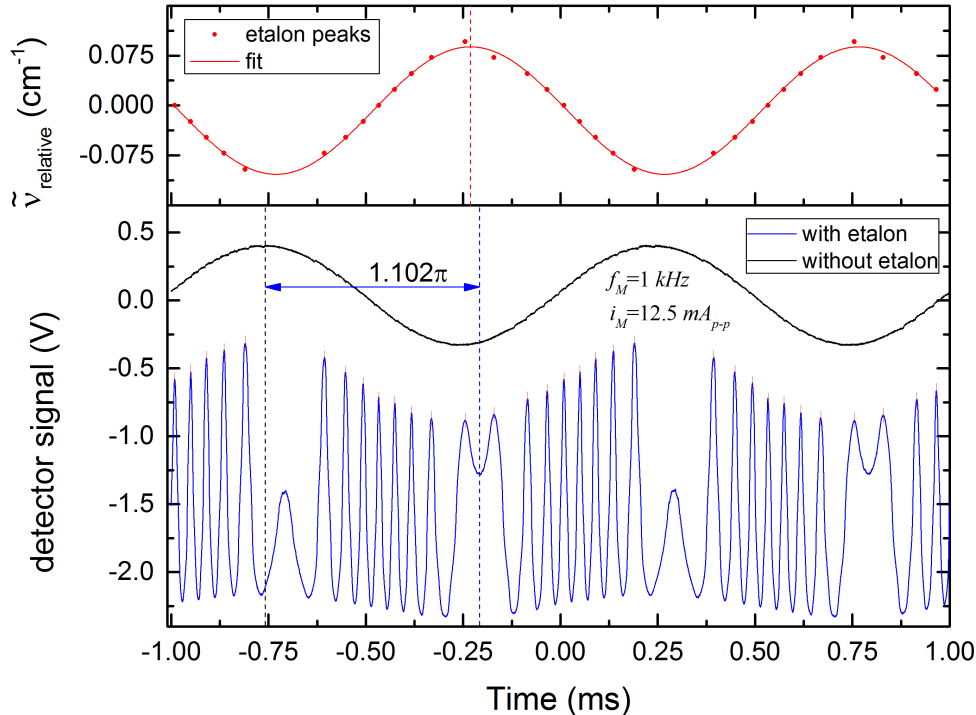
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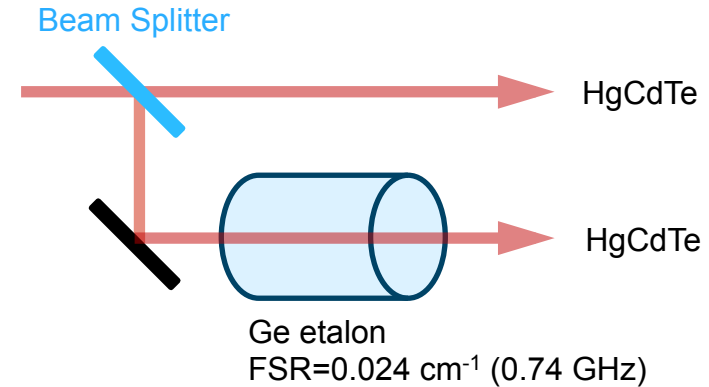
Characterization of the diode laser for WMS

WM-IM phase shift



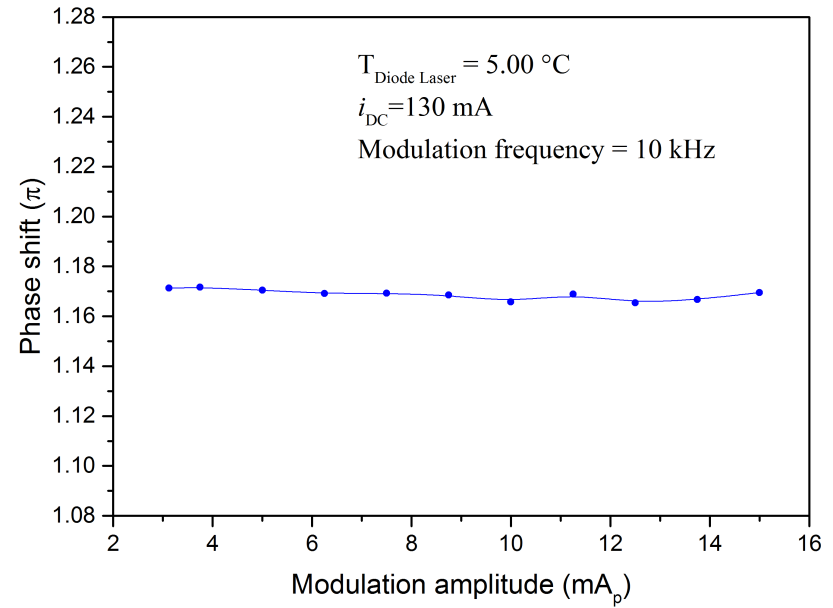
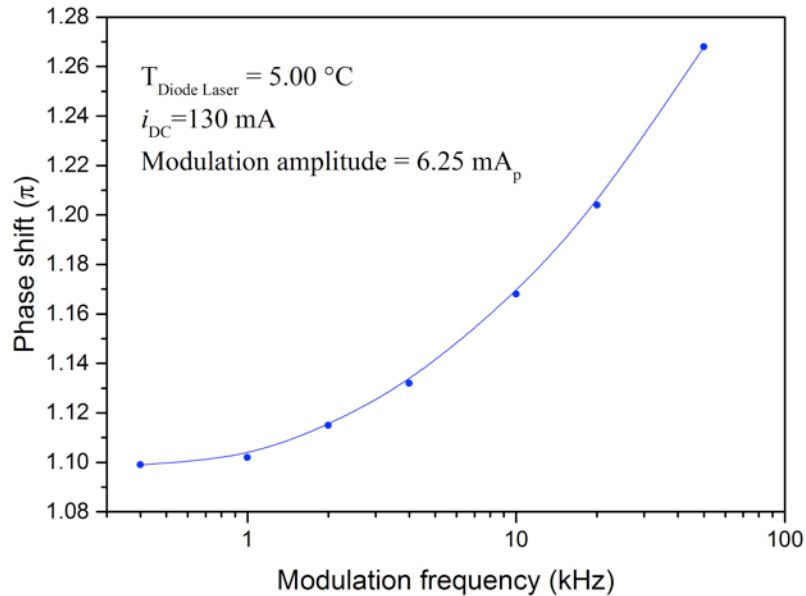
To the lowest order:

- Laser intensity varies inversely with laser frequency (i.e. phase shift = π)



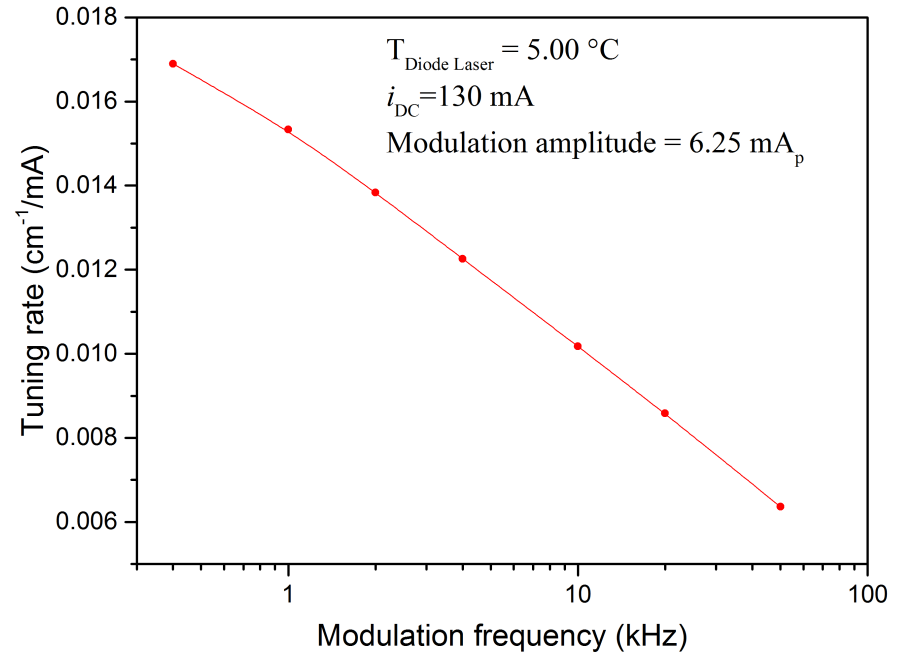
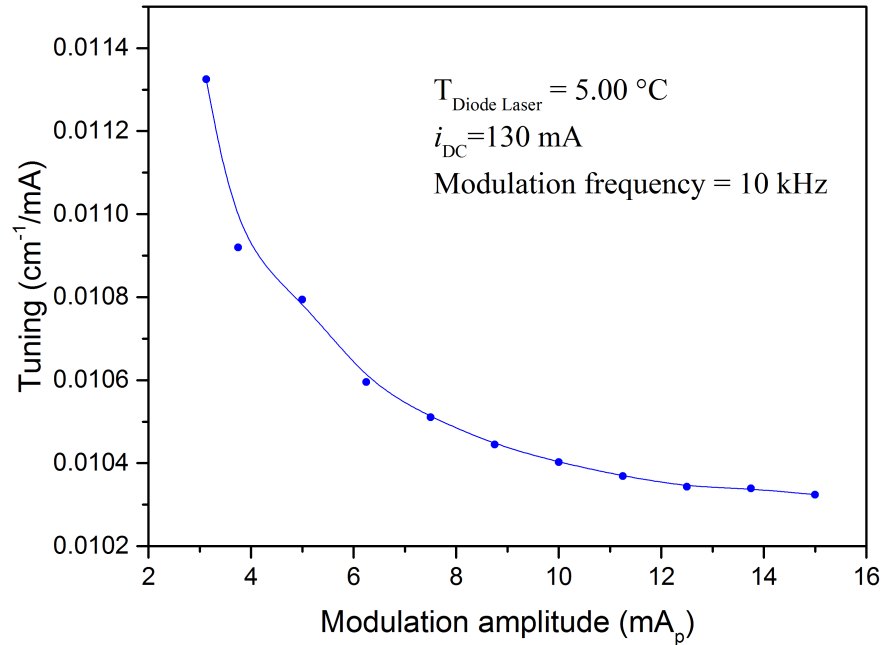
Characterization of the diode laser for WMS

WM-IM phase shift



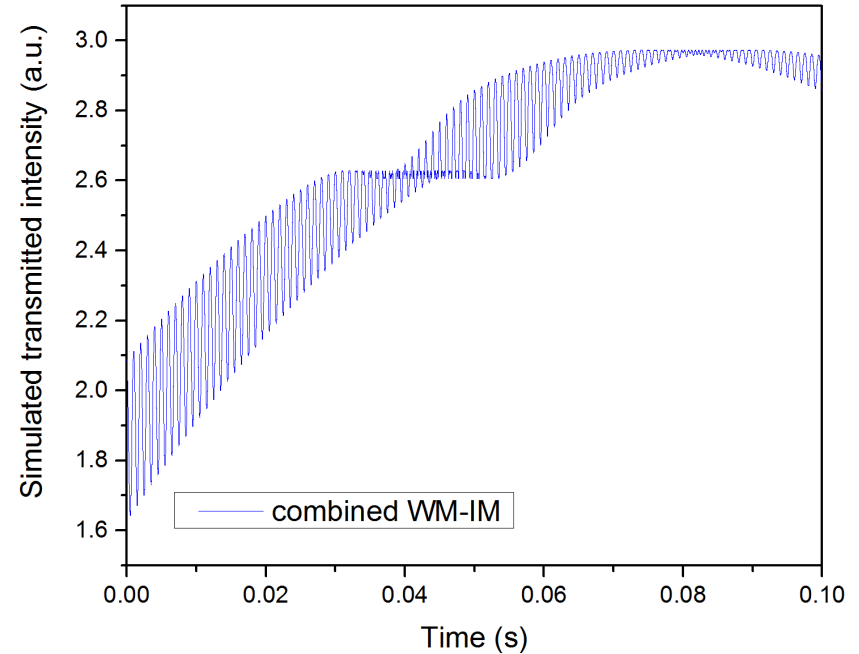
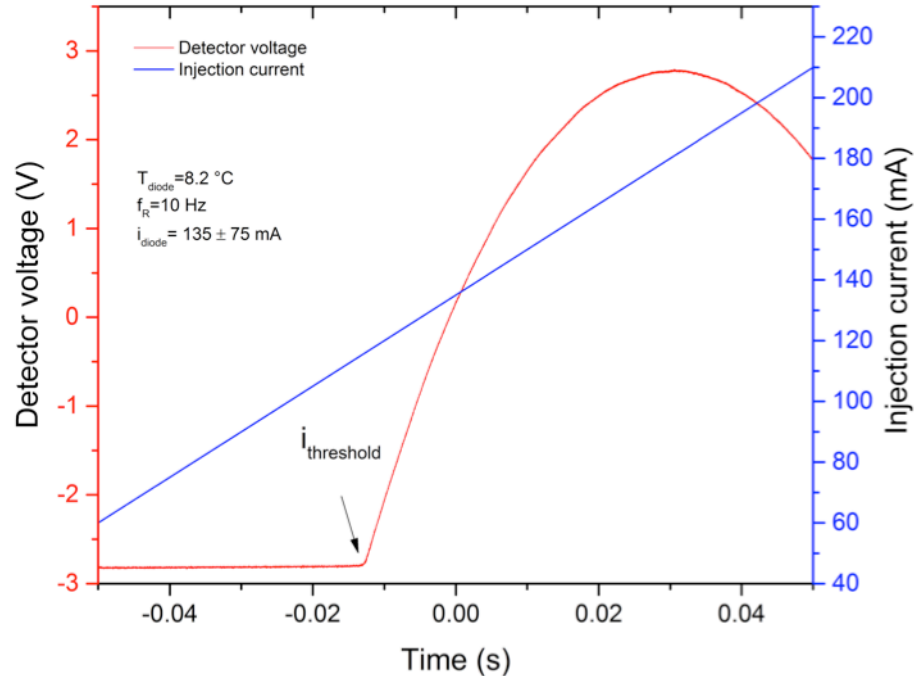
Characterization of the diode laser for WMS

Modulation tuning rates



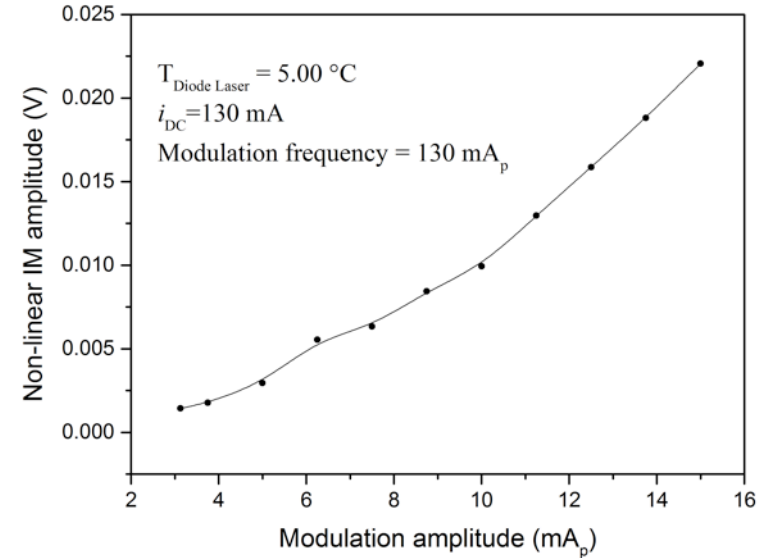
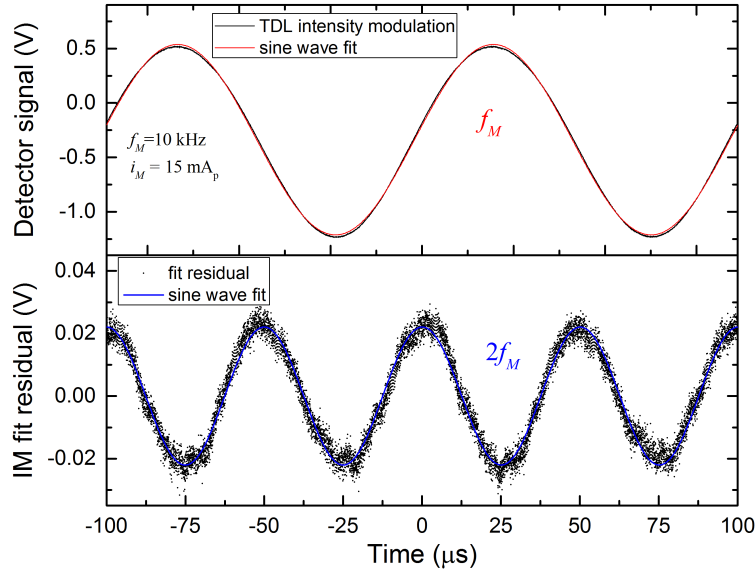
Characterization of the diode laser for WMS

Laser induced intensity modulation (IM)



Characterization of the diode laser for WMS

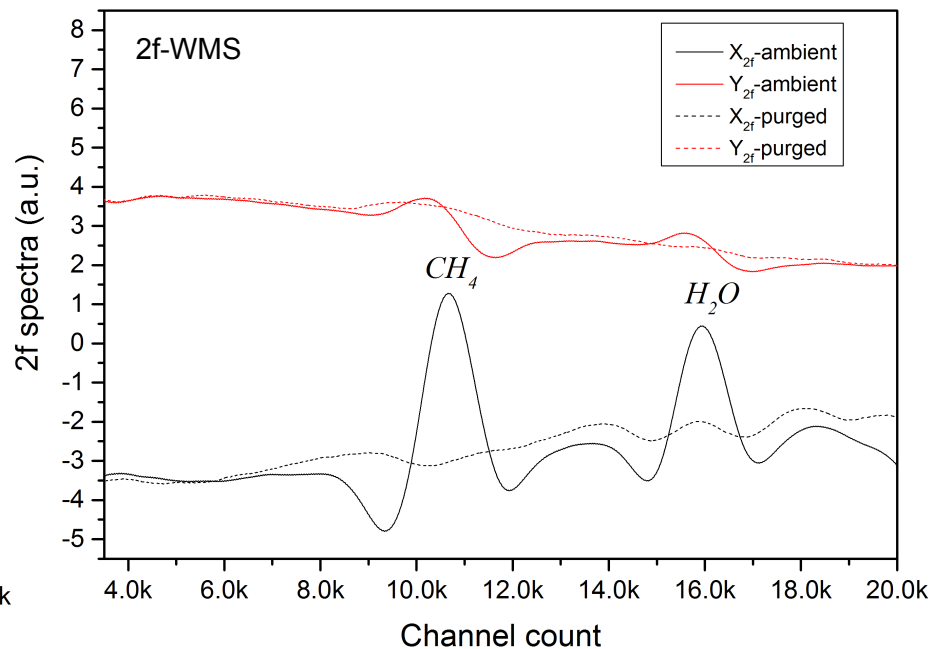
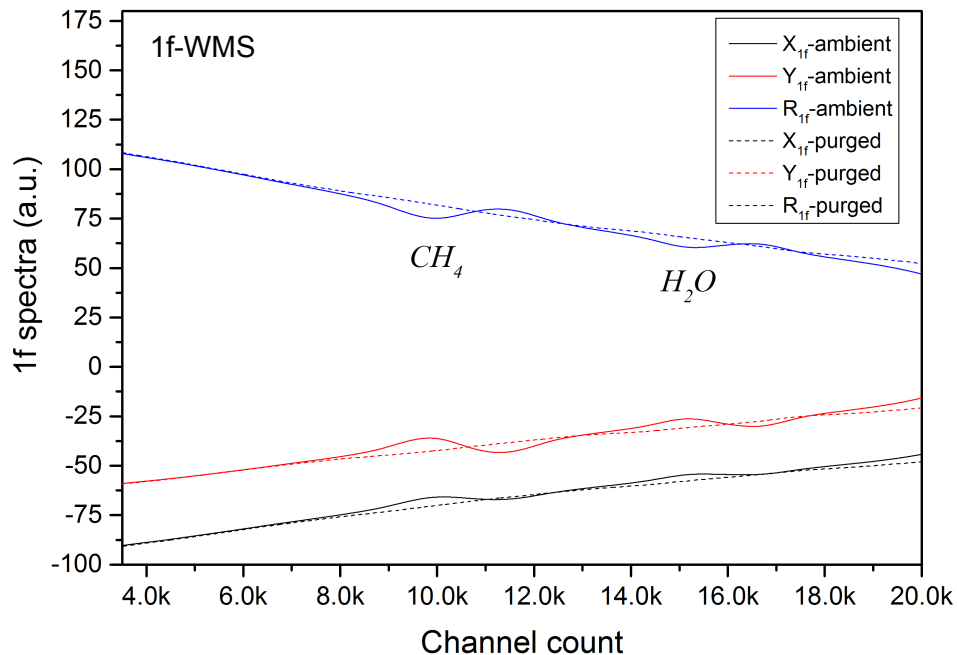
Non-linear intensity modulation



$$I(t) = I_0 [1 + i_1 \cos(\omega_M t + \phi_1) + i_2 \cos(2\omega_M t + \phi_2)] \quad \text{Linear and non-linear IM}$$

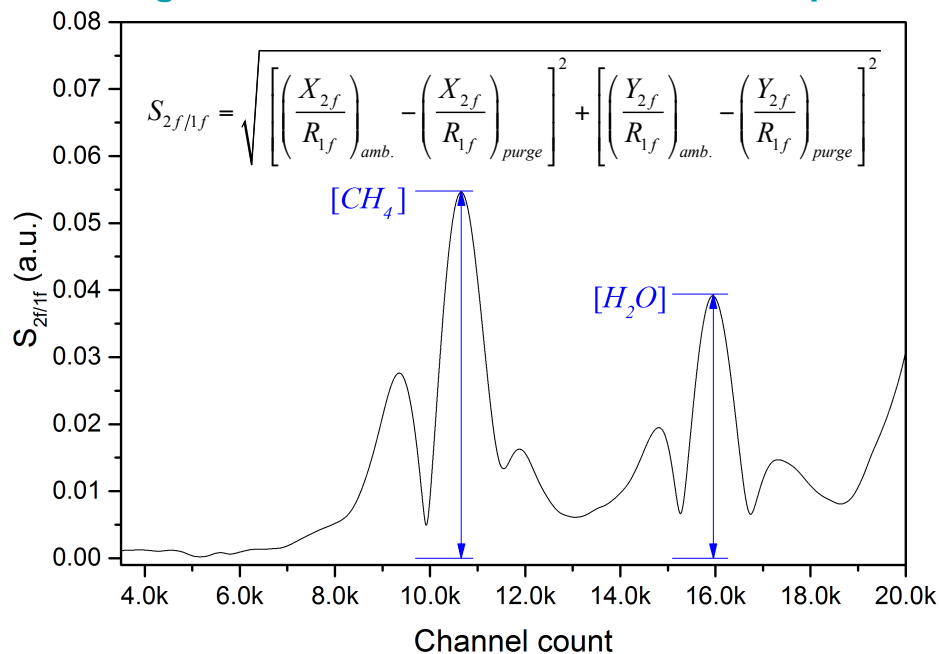
Background subtraction and 2f/1f normalization

Second and first harmonic signals with and without absorber

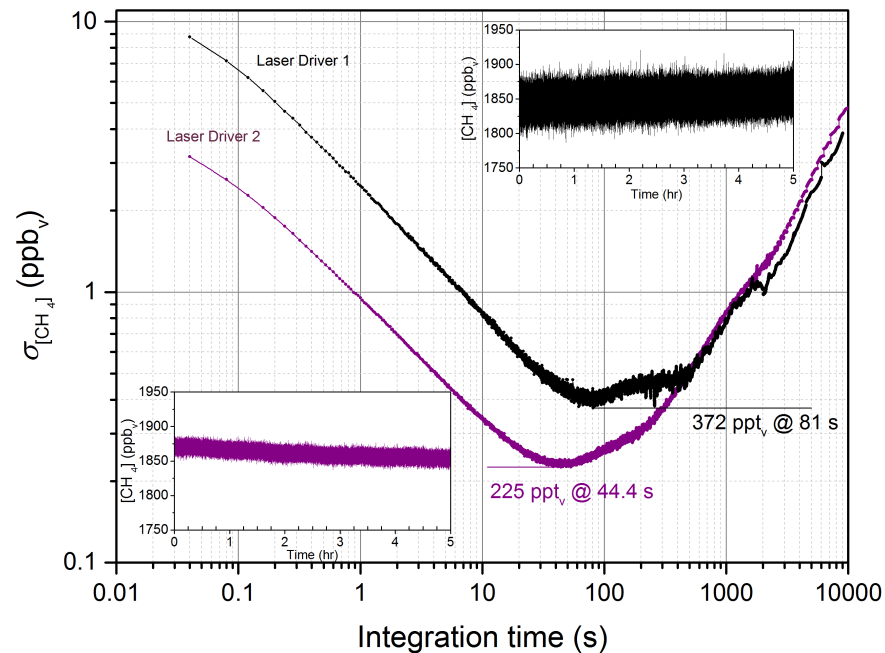


Background subtraction and 2f/1f normalization

Background subtracted 2f/1f normalized spectrum

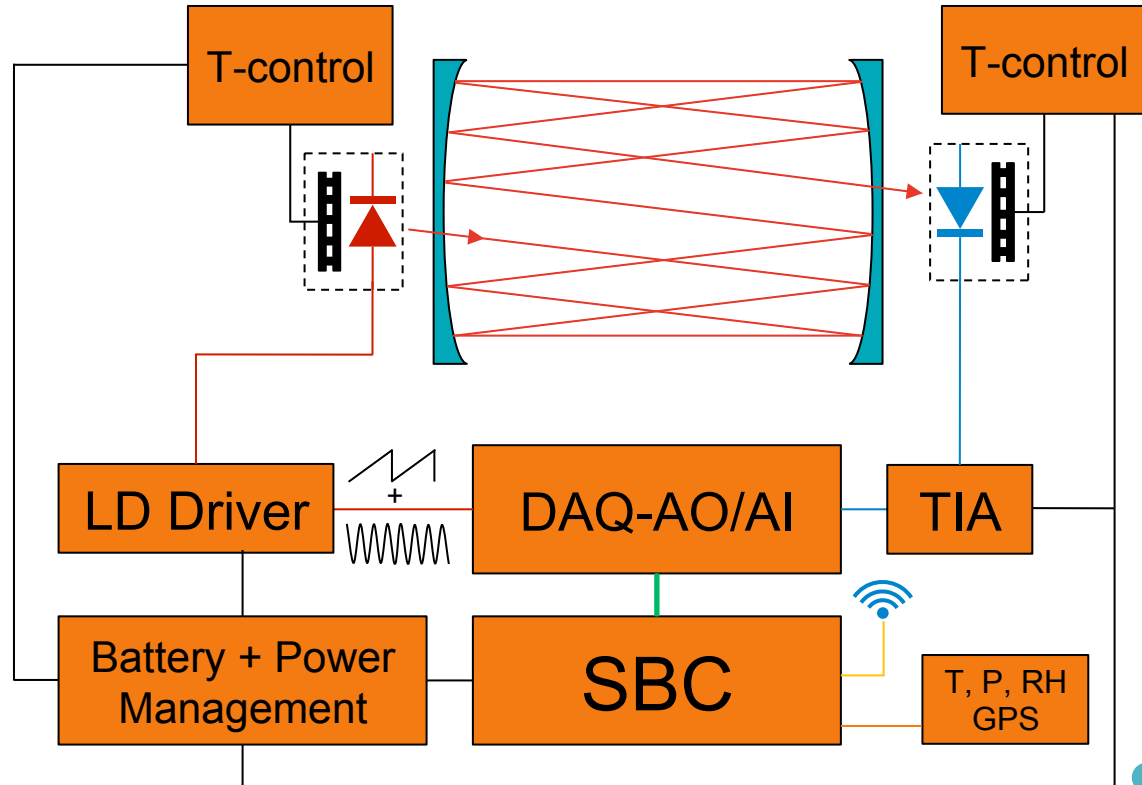


Allan deviation plot after noise reduction

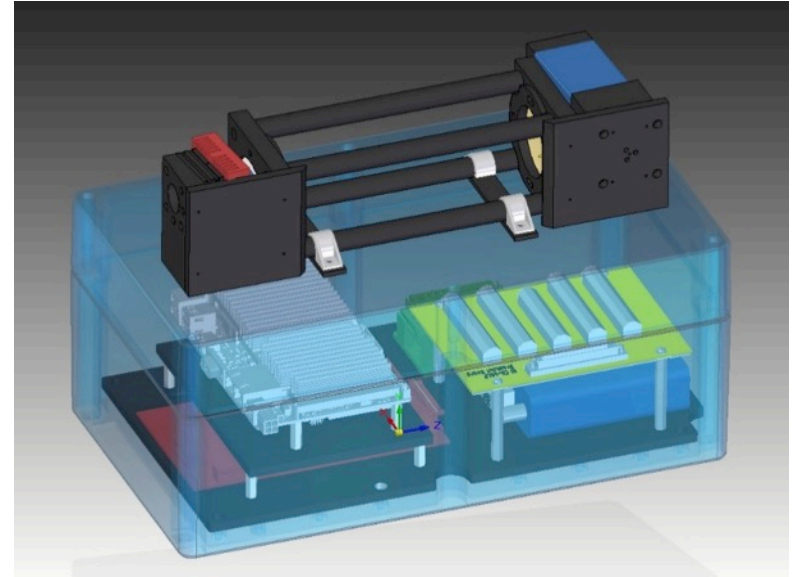
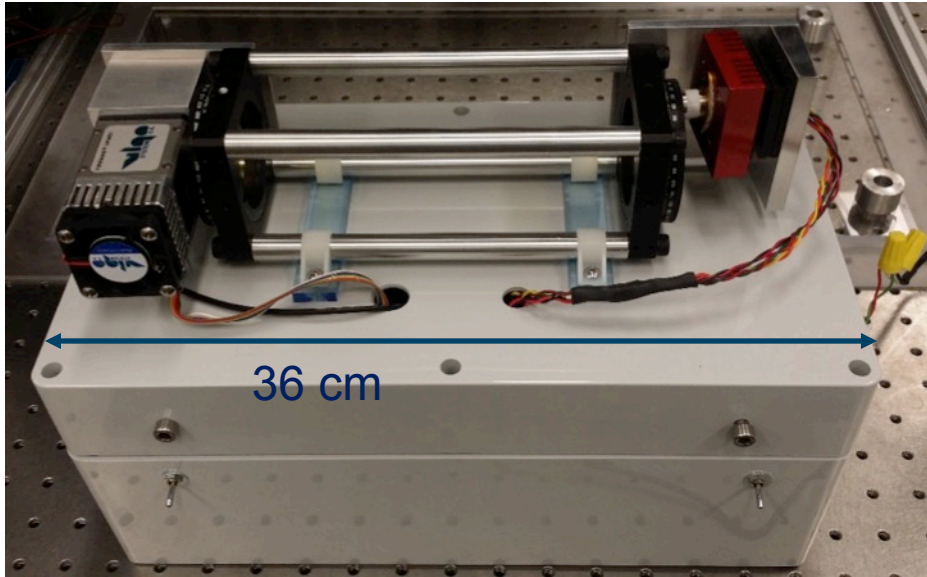


Sensor design: block diagram

- Fully programmable (LabView)
- Laser drive signal
- Software-based lock-in amplifier
- Ambient conditions sensors, GPS, Wi-Fi



Current prototype



Weight: 5 kg Power consumption: ~20 W

Future directions

Methane sensor

- Performance evaluation of the prototype in environmental chamber
- Mobile lab experiments in the field
- Further reduction in weight, size and power consumption
- UAV-based measurements over wetlands, farms, oil and gas facilities

THANK YOU

