

DFB Diode Laser Based Sensor for Isotope Ratio Detection of Methane using Continuous Wave Cavity Ring-down Spectroscopy

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I Motivation for Studying Methane Isotopes

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Motivation for Studying Methane Isotopes

1. Motivation for studying methane

On Earth

~1800 ppb

- ❖ CH_4 is a 80 times stronger green house gas than CO_2 on molecule-for molecule basis.
- ❖ Warms up atmosphere and helps form ozone.

On Mars

~10 ppb

- ❖ Peaking when it's warm in some regions.
- ❖ Might be a sign of biological activities.

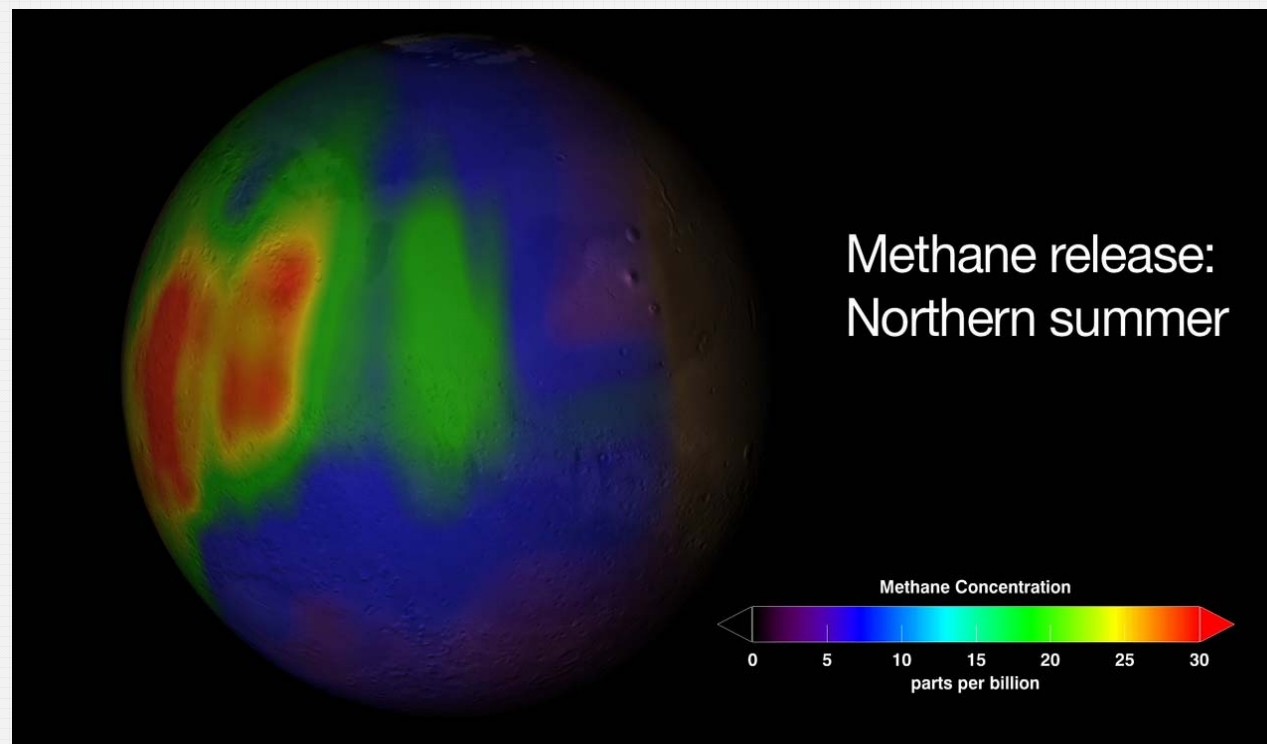


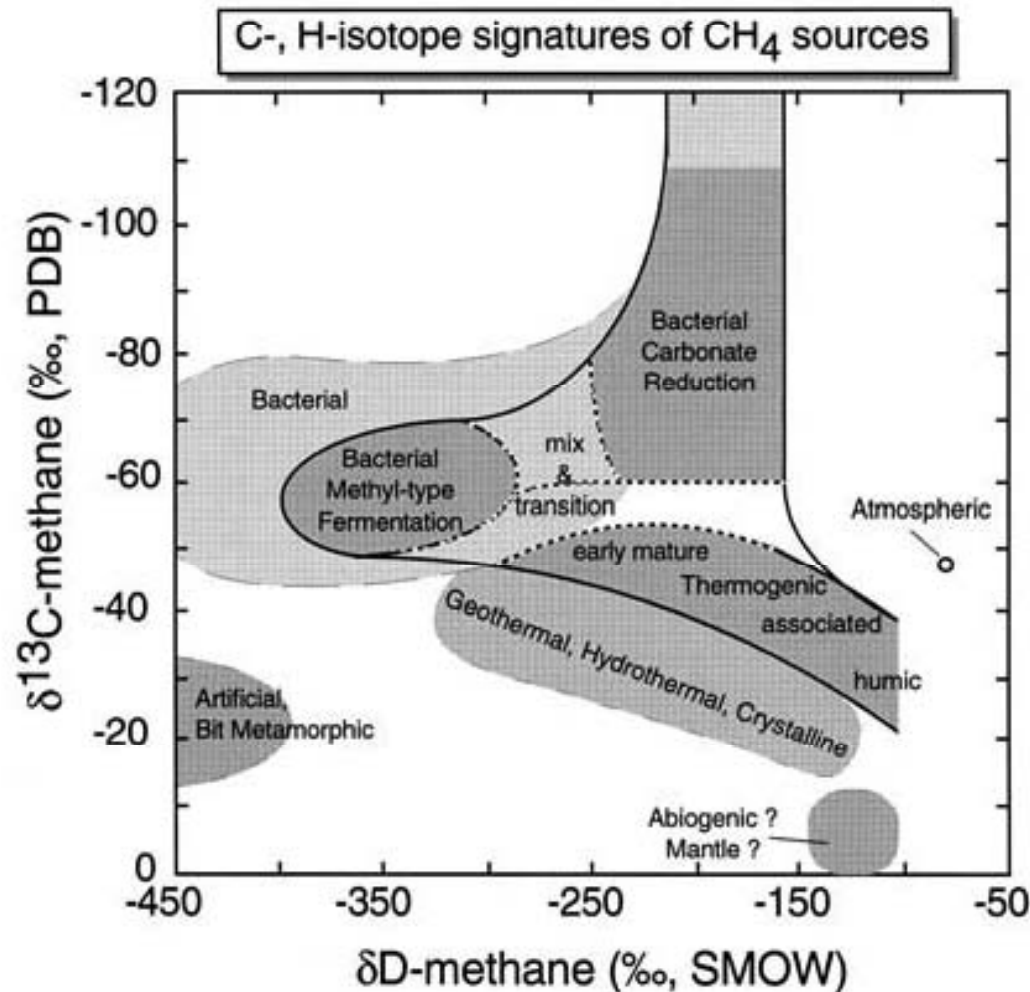
Fig 1. Concentrations of Methane discovered on Mars <1>

<1> Credit:NASA

Motivation for Studying Methane Isotopes

2. Motivation for studying methane's isotopes

Three major isotopes of methane: CH_4 , CH_3D and $^{13}\text{CH}_4$



Methane sources on earth:

Human activities

(About 2/3):

fossil-fuel extraction

rice paddies

Landfills

cattle...

Natural sources

(About 1/3):

wetlands, gas hydrates,

permafrost, termites,

oceans, freshwater bodies,

non-wetland soils,

wildfires...

Fig 2. Isotope ratio measurements on CH_4 <2>

Cavity Ring-down Spectroscopy

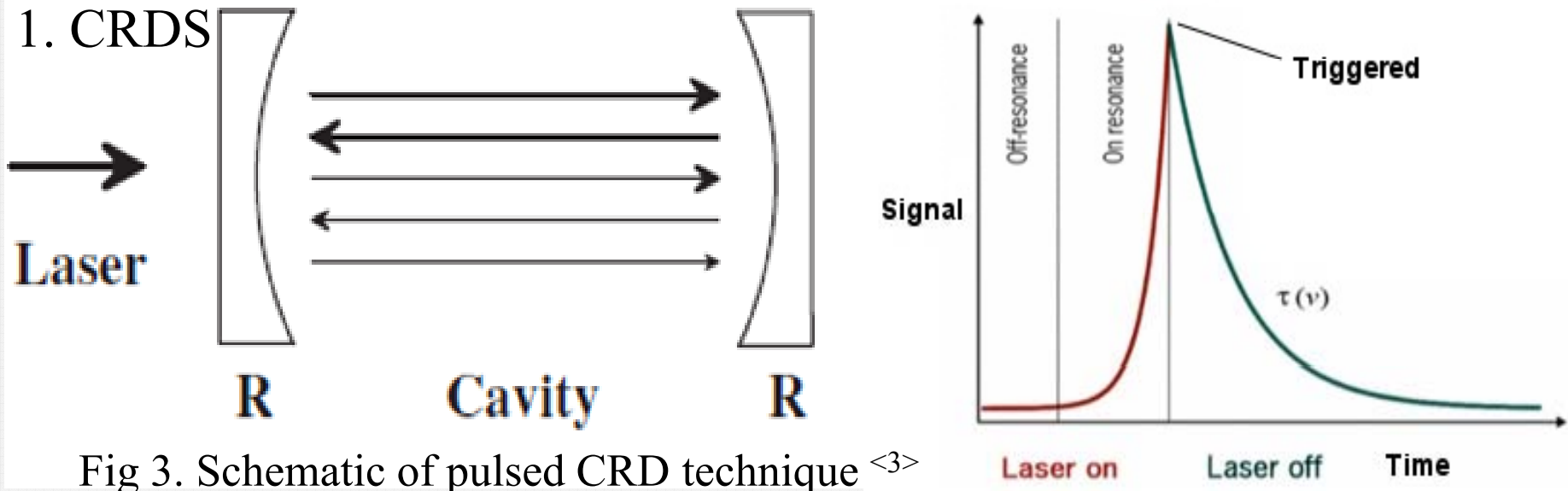


Fig 3. Schematic of pulsed CRD technique ^{<3>}

$$\frac{I}{I_0} = \exp\left(-\frac{t}{\tau}\right) \Rightarrow \Delta k(\nu) = \frac{1}{\tau} - \frac{1}{\tau_0} \Rightarrow \left\{ \begin{array}{l} \sigma(\nu) = \frac{\Delta k(\nu)}{N \cdot c} \\ N = \frac{\Delta k(\nu)}{\sigma(\nu) \cdot c} \end{array} \right.$$

Advantages of CRDS:

- High sensitivity due to long absorption length.
- Immune to intensity variations in laser.
- High throughput: individual ring down events occur on the millisecond time scale.

Cavity Ring-down Spectroscopy

2. Experimental Setup

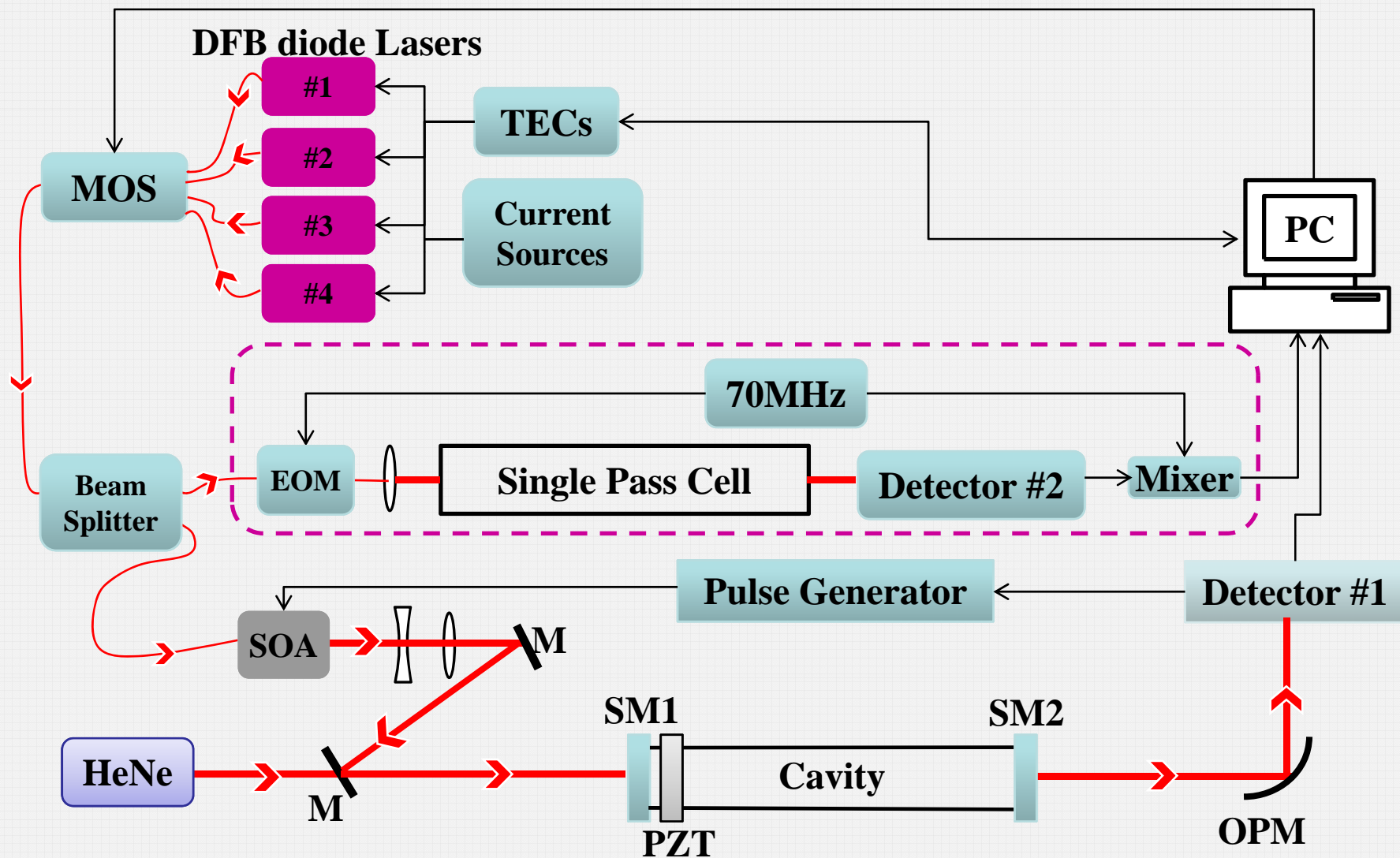


Fig 4. Schematic of Experimental Setup

Cavity Ring-down Spectroscopy

3. DFB laser diode



- ◆ Simple and small design.
- ◆ Grating structure within the semiconductor material, to serve as the wavelength selective element and reflects light back into the cavity to form the resonator.
- ◆ Tuning is accomplished by modulating either laser current or operating temperature.

Advantages:

- Absence of any critical opto-mechanical components
- High long-term stability and reliability

Applications:

- High-resolution spectroscopy
- Laser cooling, ultra-cold atoms
- Plasma physics
- Trace gas analysis
- Combustion monitoring
- Seed laser for LIDAR measurements
- Generation of tunable CW THz radiation

1. CH₃D Measurements

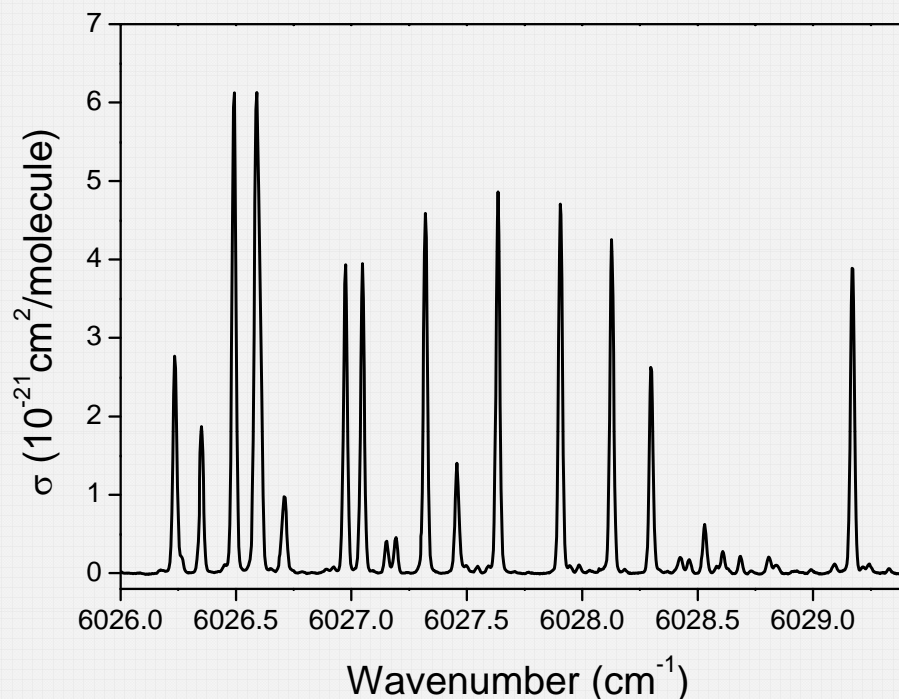


Fig 5. Cavity ring-down spectroscopy of CH₃D in ~ 8.3 Torr N₂ buffer gas.

The CH₃D number density was 5.2×10^{13} molecule/cm³.

The temperature tuning coefficient is approximately -12.5 GHz/°C, i.e. -0.42 cm⁻¹/°C.

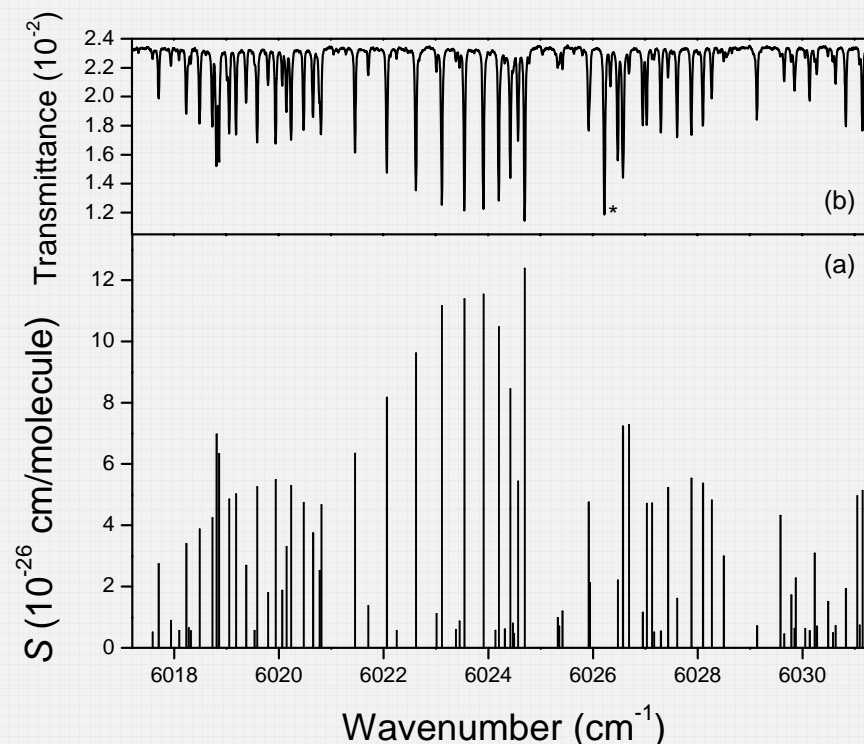


Fig 6 (a) Line strengths of CH₃D in the wavelength range of $6017.5 - 6031.5$ cm⁻¹.

The total pressure in the cavity was around 8.3 Torr. The CH₃D number densities varied between $0.5 - 2.0 \times 10^{14}$ molecule/cm³.

Fig 6 (b) CH₃D absorption spectrum from reference (room temperature, 76.7 Torr pressure, 105 m absorption path length). Resolution is 0.01 cm⁻¹. A CH₄ absorption line is marked with an asterisk.

Table 1. Part of Line strengths of CH₃D in the 6017.5 – 6031.5 and 6046.5 – 6070.0 cm⁻¹ wavenumber region, and the possible quantum numbers of the perpendicular band 2ν₄(E) transitions along with their calculated absorption positions and intensities. Not all the CH₃D lines were assigned. Measured line strengths correspond to cm per CH₃D molecule. Transitions with star (*) belong to the parallel band 2ν₄(A₁), whose absorption positions and intensities were not simulated.

Measured ν (cm ⁻¹)	Measured S (10 ⁻²⁶ cm/molecule)	Calculated ν (cm ⁻¹)	Calculated Intensity (a.u.)	Transition Assignment
6017.590	0.506			
6017.706	2.74	6017.684	0.680	^P Q ₁ (10)
6017.941	0.887	6017.767	0.633	^R Q ₀ (14)
6018.098	0.553			
6018.235	3.40	6018.229	2.195	^P Q ₁ (9)
6018.279	0.644			
6018.322	0.541			
6018.488	3.87	6018.586	0.990	^R Q ₀ (13)
6018.731	4.24	6018.730	2.673	^P Q ₁ (8)
6018.811	6.98	--	--	^Q R ₀ (4) *
6018.858	6.34	--	--	^Q R ₁ (4) *
6019.053	4.85	--	--	^Q R ₂ (4) *
6019.184	5.02	6019.185	3.099	^P Q ₁ (7)

1. CH₃D Measurements

Pressure-Broadening Coefficients

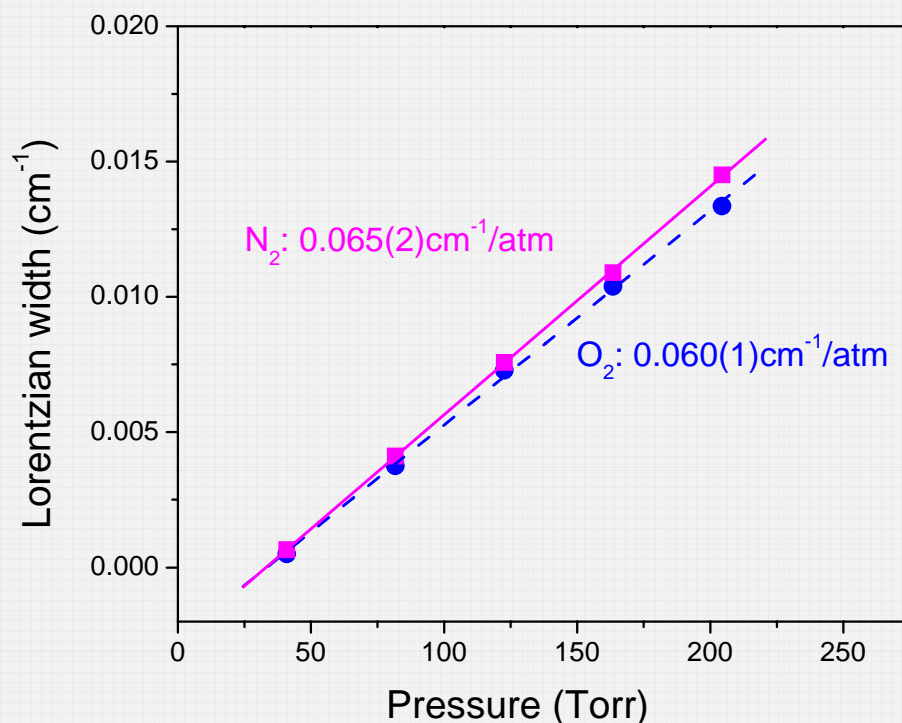


Fig 7. The N_2 (square and solid line) and O_2 (circle and dash line) pressure-broadening coefficients (HWHM) of the CH_3D absorption line at 6017.941 cm^{-1} . The scattering symbols are measurements and the lines are linear fits. The slope of the linear fit is defined as pressure-broadening coefficient for HWHM.

Pressure-Shift Coefficients

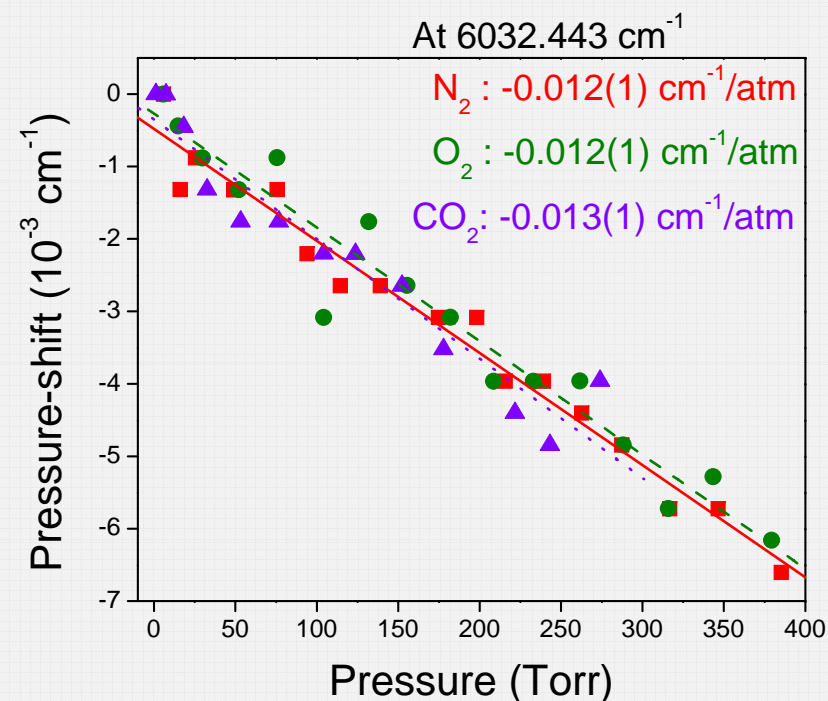


Fig 8. The N_2 (square and solid line), O_2 (circle and dash line), and CO_2 (triangle and dotted line) pressure dependence of the CH_3D absorption peak at $\sim 6032.443 \text{ cm}^{-1}$. The scattering symbols are measurements and the lines are linear fits. The slope of the linear fit is defined as pressure-shift coefficient.

2. CH₂D₂ Measurements

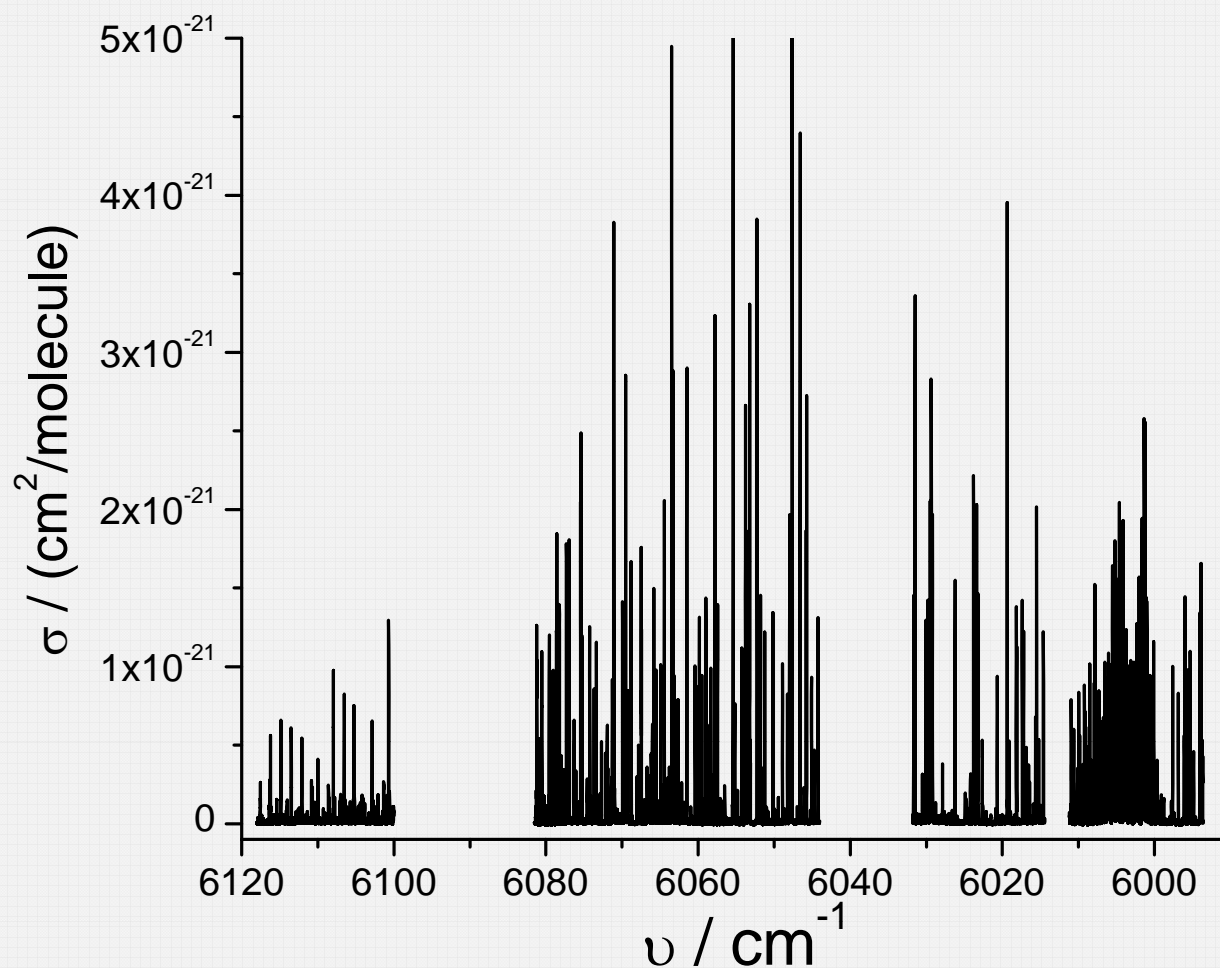


Fig 9. CRDS of CH₂D₂ absorption spectra in 16 Torr N₂ buffer gas measured by 6 different DFB diode lasers. The CH₂D₂ number density was 9.87×10^{13} molecule/cm³.

3. Electronic Feedback for locking lasers

Why do we want to lock the laser:

1. Reduced absorption cross section error $\sigma(\nu) = \frac{\Delta k(\nu)}{N \cdot c}$
2. Improved cavity transmission $T_c = \frac{T^2}{(1-R)^2} \cdot \frac{1}{1 + 2\pi\tau \cdot \Delta\nu}$

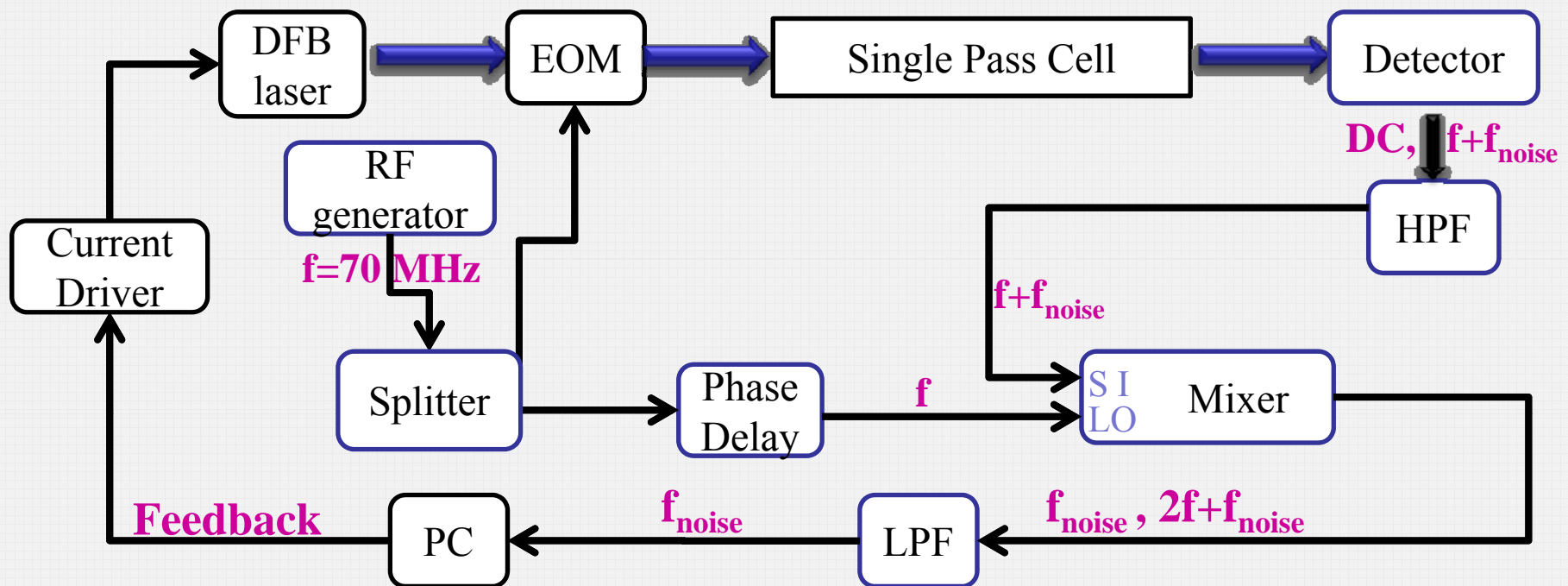


Fig 10. Schematic of 1f WMS feedback system

3. Electronic Feedback for locking lasers

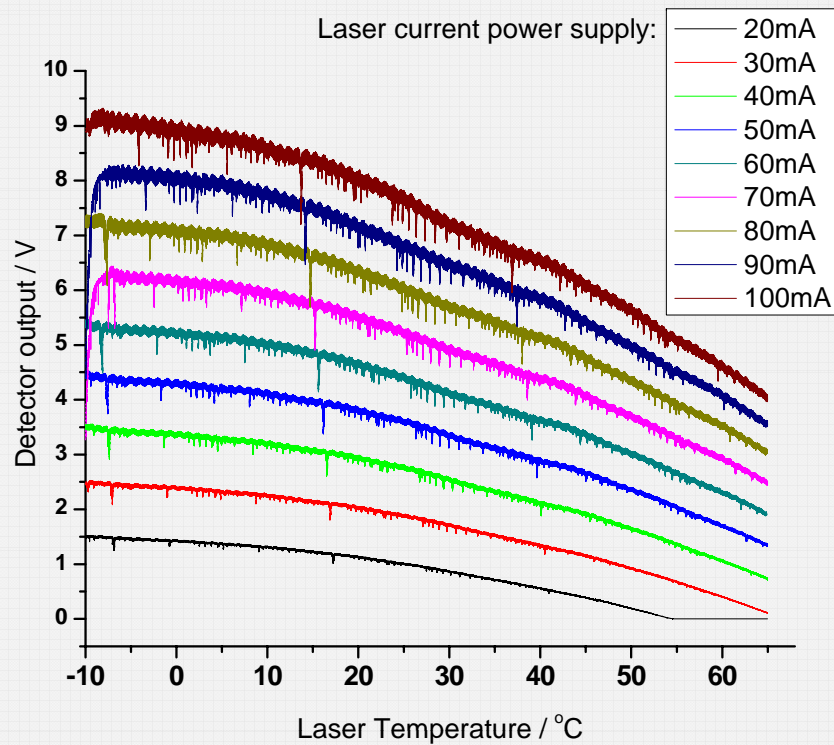


Fig 11. Detector output voltage of single pass cell filled with ~ 8 torr 98% CH_3D at different laser driver current

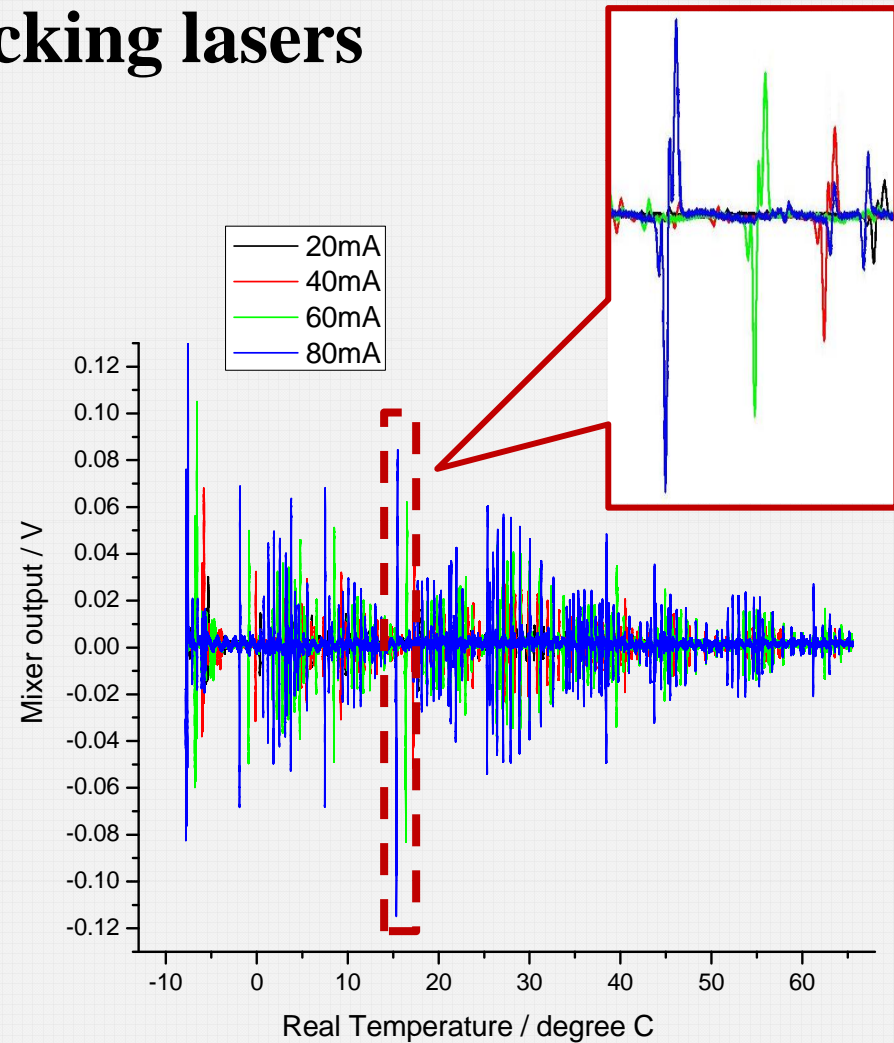


Fig 12. Mixer output variance with laser temperature tuning at different laser driver current

3. Tuning PID control

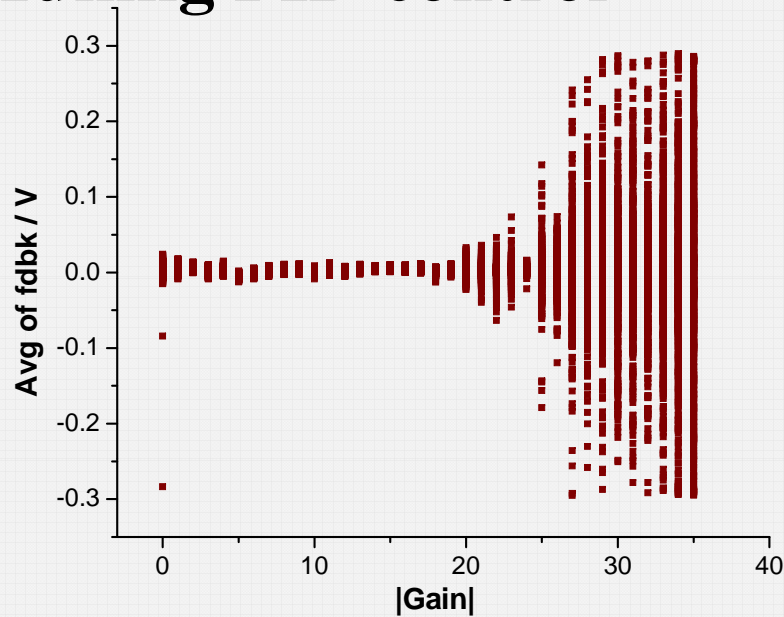


Fig 13. Feedback signal jitter with different PID total gain

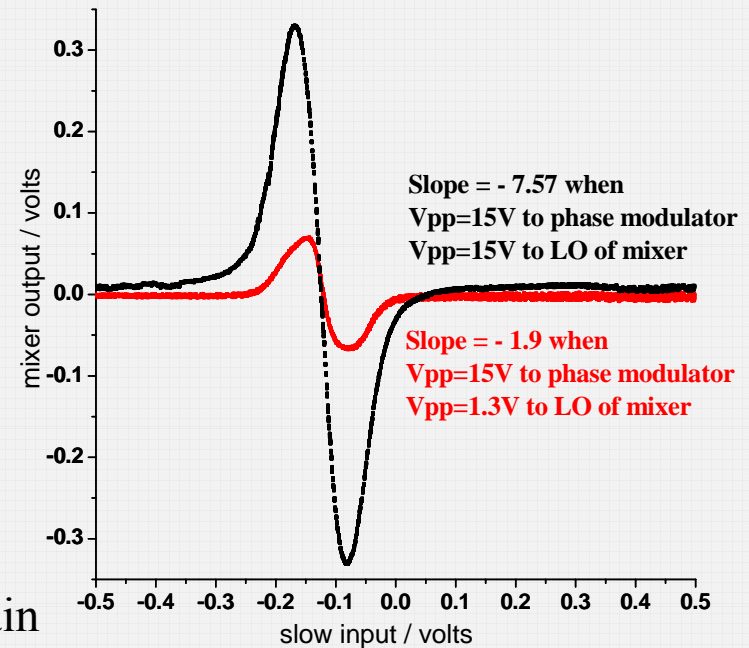


Fig 14. Examples of how modulation amplitudes affect the slope mixer output vs slow input.

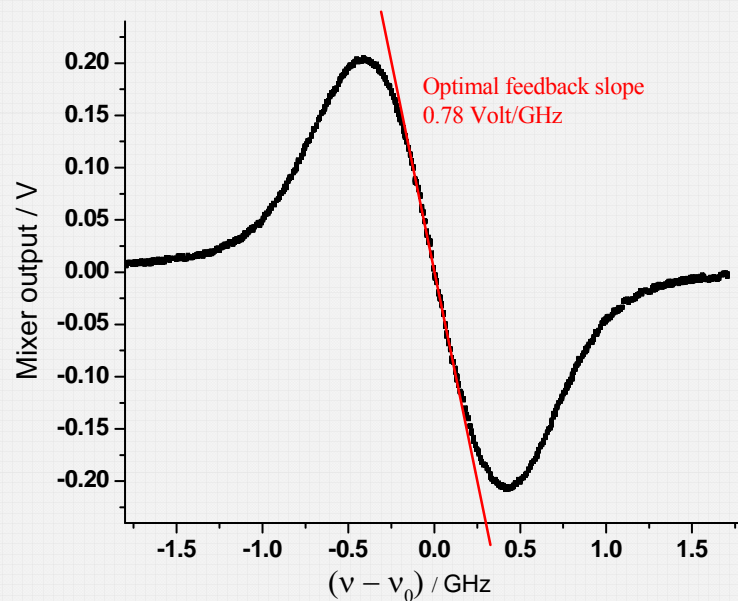


Fig 15. Optimal modulation amplitude

3. Tuning PID control

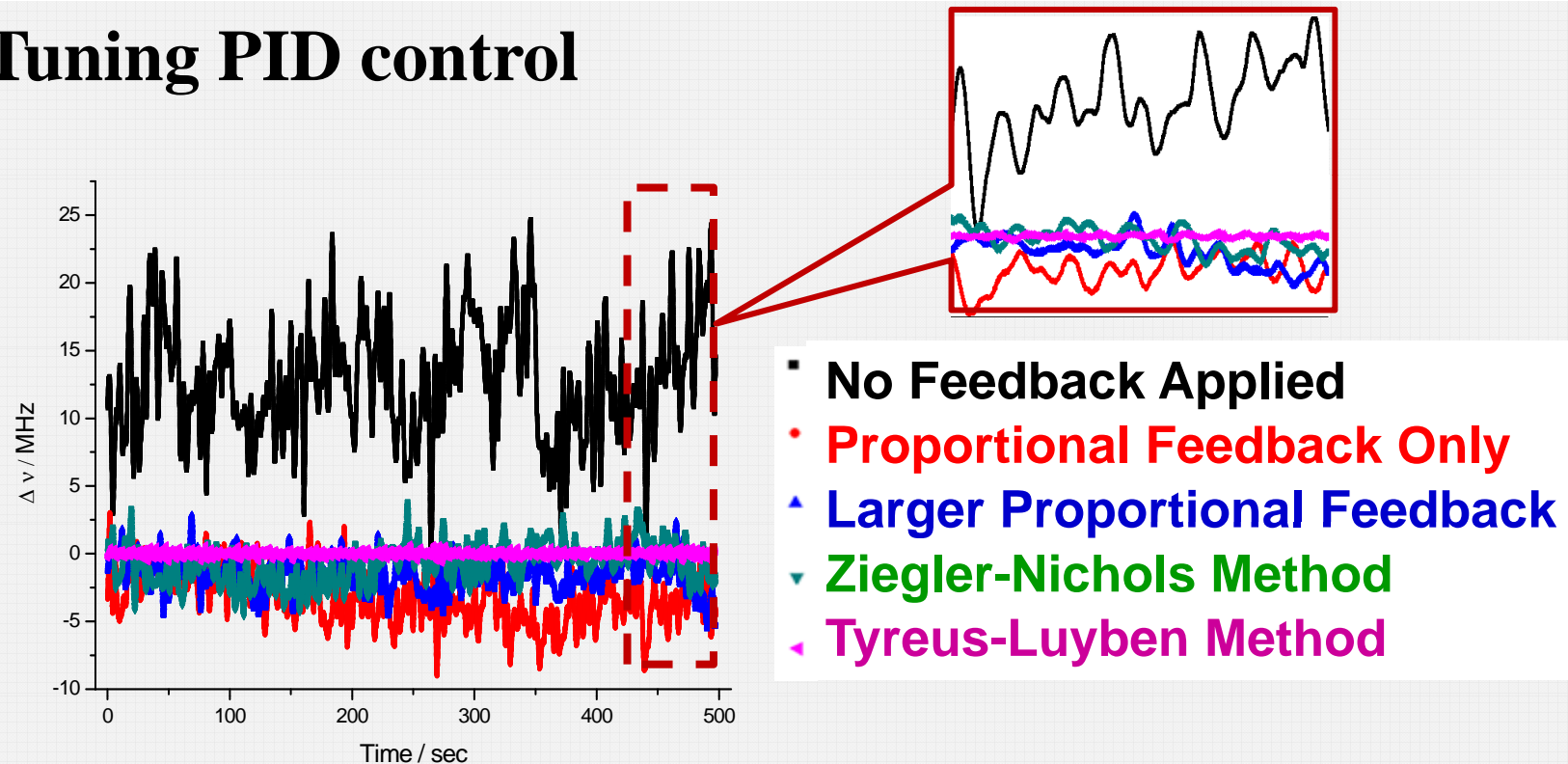


Fig 16. Comparison of laser jitter when using feedback with different PID parameters

PID parameters	Mean / MHz	Standard Deviation / MHz
No feedback applied	12.75845	4.3672
Proportional feedback only	-3.25909	1.93233
Larger Proportional feedback	-1.46707	1.25413
Ziegler-Nichols Method	-0.77357	1.44293
Tyreus-Luyben Method	-0.00046	0.18393

4. Temperature Tuning System

(1) Previous fixed current method

U is controlled by D/A and PC.

Temperature can be get from Steinhart Equation

$$1/T = C_1 + C_2 \ln(R) + C_3 [\ln(R)]^3$$

Where $C_1 = 1.1292\text{E-}3$, $C_2 = 2.3411\text{E-}4$,

$C_3 = 8.7755\text{E-}8$, $I = 100 \mu\text{A}$, $R = U / I$.

(2) New voltage divider method

Instead of a fixed I, we used a voltage divider, with the thermistor in series with fixed resistor R', and both thermistor R and fixed resistor R' are driven by a fixed voltage source U'.

The output voltage will be proportional to R'. $R = R' \times U / (U' - U)$

4. Temperature Tuning System

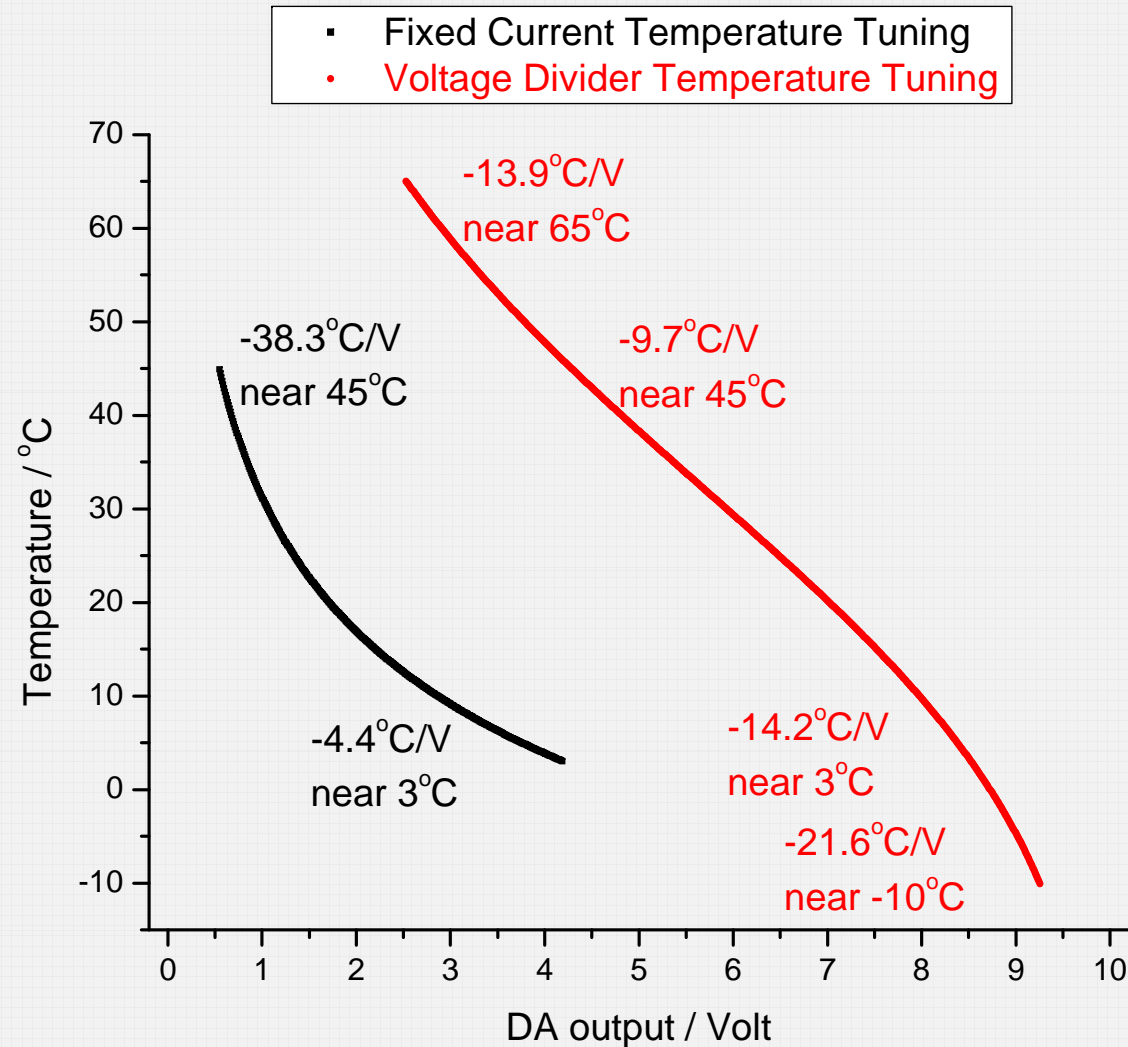


Fig 17. Comparison of two methods of temperature tuning

Future Plans

- I. Super narrow linewidth DFB lasers
- II. Electronic feedback and temperature tuning on all lasers
- III. Implement cavity locking with predicted wavelength shift

Acknowledgements

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