Toward Precision Mid-Infrared Spectroscopy on the OH Radical

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Our Experiment

- Precision measurements of OH $X^2Π_{3/2}$, $ν' = 2 \leftrightarrow ν'' = 0$ at $2 \times 2.87 \, \mu m$ (3580 cm$^{-1}$, 104 THz)
- Goal: measure transition frequency at the level of the natural linewidth $Γ = 6$ Hz ($2 \times 10^{-10}$ cm$^{-1}$)
  - Relative precision of $\sim 3 \cdot 10^{-14}$
- Transition is sensitive to possible time-variation of $m_e/m_p$
Many dimensionless physical constants whose values cannot be predicted mathematically

\[ \alpha \equiv \frac{e^2}{4\pi\epsilon_0\hbar c} \approx \frac{1}{137.035\,999\,139}(31) \text{ (CODATA 2014)} \]

\[ \mu \equiv \frac{m_e}{m_p} \approx \frac{1}{1836.152\,673\,89}(17) \text{ (CODATA 2014)} \]

Possible that these values change in time and/or space

(Uzan, Rev. Mod. Phys. 75, 403–455 (2003))
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Changes in \( \mu \) lead to shifts in rotational, vibrational transitions

\[ B \sim \mu \implies \frac{\Delta \nu_{\text{rot}}}{\nu_{\text{rot}}} = \frac{\Delta \mu}{\mu} \]

\[ \omega_e \sim \sqrt{\mu} \implies \frac{\Delta \nu_{\text{vib}}}{\nu_{\text{vib}}} = \frac{1}{2} \frac{\Delta \mu}{\mu} \text{ (but larger absolute shift)} \]
Experimental overview

- 1064nm @ 20°C
- PPLN
- 1.7 µm
- Cell
- 532 nm
- 616 nm @ -116°C
- BIBO
- 308 nm
- 776 nm
- 2.9 µm
- PPLN
- 1.7 µm
- 2.9 µm
- I₂ Cell
- 308 nm
- 616 nm
Pulse train from mode-locked laser equivalent to many CW lasers in the frequency domain

Optical frequency of every comb tooth determined by only two parameters

- $f_r$ — repetition rate of laser ($\sim 1$ GHz)
- $f_0$ — phase slip relative to envelope (between 0 and $f_r/2$)
Absolute measurement of optical frequencies

Absolute frequency of CW laser given by
\[ f_{\text{CW}} = nf_r + f_0 + f_{\text{beatnote}} \]

Arrows indicating:
- \( f_{\text{beatnote}} = f_{\text{CW}} - nf_r - f_0 \)
- \( f = nf_r + f_0 \)

- \( f_r \), \( f_0 \), and \( f_{\text{beatnote}} \) are radio frequencies, can be compared to atomic clock reference
- \( n \) is an integer
Stabilizing the comb with a 1064/532 nm laser

\[ f_{bn,1064} = f_{CW,1064} - nf_r - f_0 \]
\[ f_{bn,532} = 2f_{CW,1064} - 2nf_r - f_0 \]

From these beatnotes, we can calculate

\[ f_{bn,532} - 2f_{bn,1064} = f_0 \]
\[ f_{bn,532} - f_{bn,1064} = f_{CW,1064} - nf_r \]

Direct comparison of \( f_{CW,1064} \) and \( f_r \)
Frequency measurement of 1064-nm laser

$f_{bn,1064} = -66.4\ \text{MHz}$

$f_{bn,532} = -90.5\ \text{MHz}$

$f_r = 999.908798\ \text{MHz}$

$n = 281656$

$RBW: 1\ \text{MHz}$

$VBW: 30\ \text{kHz}$

$f_0 = 42.3\ \text{MHz}$

$f_{CW,1064} = 281\,630\,288\ \text{MHz}$
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High-precision saturated absorption lock to $I_2$ transition

- 1064-nm laser very stable at short times, but drifts
- Rubidium clock, GPS only improve stability on timescales $\gtrsim 1$ s
High-precision saturated absorption lock to I$_2$ transition

- 1064-nm laser very stable at short times, but drifts
- Rubidium clock, GPS only improve stability on timescales $\gtrsim 1$ s
- Iodine reference bridges gap between short and long timescales

Döringshoff et al., “High performance iodine frequency reference for tests of the LISA laser system”, EFTF-2010
High-precision saturated absorption lock to I$_2$ transition

- Low-temperature cold finger (reduces pressure shift)
- Frequency-shifted pump (eliminates shifts from back reflections)
- Active stabilization of beam pointing
Modulation transfer spectroscopy of R(56) 32-0 transition

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Singly-resonant CW OPO pumped at $\lambda_p = 1064$ nm, 15 W.
- For OH transition, $\lambda_s = 1690$ nm, $\lambda_i = 2870$ nm
- Small amount of $\lambda_p + \lambda_i$ produced / measured using comb
  - $\left( \frac{1}{\lambda_p} + \frac{1}{\lambda_i} \right)^{-1} = 776$ nm
- High bandwidth (200 kHz) piezo mirror for stabilising idler
- Idler tunable from 2400 to 4000 cm$^{-1}$ (4.17 – 2.5 $\mu$m)
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PPLN
Diode laser

Cutoff for laser diodes near 633 nm
Lower temperature $\rightarrow$ larger bandgap, shorter wavelength

Diode laser dewar

Grating feedback, external cavity diode laser attached to the bottom of liquid N$_2$ dewar in vacuum
Single mode operation at 615 nm

~100 mW output power
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PPLN
My Group

John Moore-Furneaux
Visiting professor Jan.–Dec. 2015

Arthur Fast
PhD student since Feb. 2015
Tuning curves for the PPLN crystal

- Poling periods (from top to bottom): 28.5 µm, 29.0 µm, 29.5 µm, 30.0 µm, 30.5 µm, 31.0 µm, and 31.5 µm
- Coarse tuning by changing poling period
- Fine tuning by adjusting temperature