Fiber comb lasers for spectroscopy

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Overview

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  QCL spectroscopy
• Summary
**Frequency combs & standard molecular spectroscopy techniques**

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**Frequency combs for molecular spectroscopy:**

- only competes with MS in terms of cost; to compete with MS:
  - **need broadband** mid IR measurements
  - **need high spectral resolution** ($\approx 10 – 100$ MHz for unambiguous gas analysis)
  - **need high sensitivity** ($=sufficient$ spectral brightness)

For scientific applications:
- narrow linewidth allows proper evaluation of lineshapes, allowing collision effects to be accounted for.
How much IR power do we need?

**Glowbar** spectral power density = 0.01 mW/nm
(adapted for 1 GHz resolution FTIR, inversely proportional to FTIR resolution)

**Incoherent fiber SC** spectral power density = 1 mW/nm
(at 3000 nm with a fluoride fiber)

**Comb technology** can give us around 100 times better sensitivity than incoherent sources with same spectral power

**Comb technology should produce** 100 times better sensitivity to make the additional expense of combs tolerable

Coherent IR laser source should have:
spectral power density > 1 mW/nm

Ref. S.P Davis et al., Fourier Transform Spectrometry, Academic Press(2001)
Spectral power density of current coherent mid IR sources

- **PPLN**
- **OPGaAs**
- **GaSe**
- Chalcogenide fiber

**Graph:**
- **Red line** = incoherent fiber supercontinuum sources
- **Black line** = glow bar with 1 GHz res. FTIR

**Legend:**
- OPO
- DFG
- SC

**X-axis:** Wavelength (µm)
**Y-axis:** Spectral power density (mW nm⁻¹)
Why mid-IR?

Comparison of molecular absorption linestrength:

Mid-IR combs promise higher sensitivity for molecular sensing.
Tm fiber based comb sources
Tm-doped Fiber Properties

- Efficient cladding pumping at 790 nm → excellent power scaling capability (1 kW cw-power demonstrated)
- Large bandwidth & gain → ideal for fs-pulse amplification

Moulton et al., JSTQE 15, 85 (2009)

Jackson and King, JLT 17, 948 (1999)
400 MHz Tm soliton comb oscillator

Low RIN 1564 nm 2 W pump

Intracavity dispersion -17,000 fs²

Time bandwidth product 0.31, soliton pulse

Graphene Modulator

WDM

TSF

DCF

SMF

PBS

QWP

SA

FROG Intensity
Retrieved Phase

retrieved spectrum
retrieved phase
measured spectrum

Intensity [a.u.]

Delay [fs]

Wavelength (nm)

Phase [rad]

Intensity (a.u.)

Phase (rad)

FWHM 58fs
400 MHz Tm soliton amplifier

Low RIN 1564 nm 2 W pump

Graphene Modulator

Ref.: J. Jiang, “500 MHz, 58fs highly coherent Tm fiber soliton laser”
CLEO 2012 PDL, paper CTh5C.4.
Graphene Modulator design

C.-C. Lee et al., Optics Express 20, 5264 (2012)

Nobel Prize in Physics for 2010 to Andre Geim & Konstantin Novoselov

C.-C. Lee et al., CLEO (2012), paper JTu1M.3.
Graphene loss modulator for fast FCEO control

Large effect, since gain cannot follow

Small effect at DC due to gain response
locking performance (500MHz comb)

FT-IR measurement

OSA measurement

Coherent SC generated with Tm fiber system

Tm comb lock to SF diode laser

out of loop beat with SF Yb fiber laser

fceo lock
The Future of Mid-IR Frequency Conversion

Quasi-Phase Matched (QPM) GaAs:

Advantages of QPM GaAs
Large nonlinear coefficient (94 pm/V)
Non-critical quasi-phase-matching & long interaction lengths for reduced cw threshold (up to 70 mm)
High transparency out to 15 µm
High thermal conductivity (55 W/mK)
Low absorption losses (<0.005 cm$^{-1}$)

mid-IR generation with OP-GaAs OPO

Collaboration with:
Nick Leindecker & Konstantin L. Vodopyanov (CREOL) & Peter G. Schunemann (BAE Systems)

Tm fiber pump

OPGaAs crystal

2.6 – 6.1 µm output

Threshold pump energy ≈ 100 pJ,
Compatible with multi-GHz rep. rates

How to lock OPO?

- Use spurious doubled signal output from OPO for OPO cavity length control
- Rep. rate and $f_{\text{ceo}}$ of OPO automatically locked
OPO lock performance

- cavity length changes move the cw laser + OPO beat
- feedback beat to OPO mirror position (5 kHz bandwidth)
- phase lock to pump
  - linewidth below 100 mHz
  - >80% power in carrier
  - <500 mrad cumulative phase noise
Frequency combs assisted QCL molecular spectroscopy

- QCL line narrowing
- Precision frequency scanning with QCLs
- Application to molecular spectroscopy
Independent non-linear mixing schemes

1. Comb down-conversion (DFG scheme)

\[ f_{\text{DFG}} = f_n - f_m \]

\[ f_{QCL} \]

\[ f_m = f_{\text{CEO}} + mf_{\text{REP}} \]

\[ f_m = f_{\text{CEO}} + nf_{\text{REP}} \]

2. CW-laser up-conversion comb (SFG scheme)

\[ f_{\text{SFG}} = f_m + f_{QCL} \]

Gatti et al. *Optics Express*, 2011, 19, 17520
Comb assisted spectroscopy: Tm fiber comb & 9 µm QCL

1.95 µm

Tm:fiber

\( \lambda/2 \)

PBS

SC

Delay

SF 1.6 µm

AgGaSe\(_2\)

LO

Servo

Current Driver

QCL

MCT

NH\(_3\)

9.1 µm
Quantified Line-Narrowing

- Observed line narrowing from 520 kHz to 25 kHz in 1 ms.
- Possible to line narrow 2 MHz bandwidth mid-IR sources to kHz level using GPS-referenced combs!
- Narrower line widths possible with optically referenced combs.

1\textsuperscript{st} Application: Ammonia spectroscopy
Ammonia Spectroscopy

ν₂ band of Ammonia

Current Hitran line-center accuracy ~ 3 MHz.
Rapid, comb-assisted spectroscopy enables highly reproducible scans over multiple lines.

Tm oscillator can tune 5.5 GHz or 0.2 cm⁻¹ continuously.
Doppler width can be measured to give the thermodynamic temperature if molecular mass ($M$) and Boltzmann constant ($k_B$) is known.

\[
\Delta \nu_D = \frac{\nu_{\text{line}}}{c} \sqrt{2 \ln 2 k_B \frac{T}{M}}
\]

Precise knowledge $\nu_{\text{line}}$, lineshape, and stability of $T$, gives $k_B$. 

Precision Spectroscopy and Thermometry
Thermometry

Precision Thermometry History using NH$_3$:

• Previously done with single lines of CO$_2$ lasers
  • with huge datasets to reduce the statistic uncertainty

Multi-line fitting

• Remove uncertainty of fit
• Three closely spaced lines
• Same pressure and temperature
Thermometry: Line shape

Measured temperature dependent on true line shape. Quantify accuracy of line shape by looking at $T_d/T_{\text{measured}}$.

- be independent of pressure
- “= 1”

Linear scaling law ($\nu_D \sim \nu_0$)

- Reduces the statistical uncertainty on $T_{\text{gas}}$
- Voigt Line shape: $T_d/T_{\text{measure}}$ has negative slope across a pressure range.
  - Demonstrates comb-assisted spectroscopy’s accuracy in observation of collisional effects.
  - Voigt profile is insufficient to measure temperature.
- Speed dependent Voigt profile has 0 slope across pressure range.
How about phase-locking of tunable external cavity QCLs?

Line narrow QCLs ~120 cm\(^{-1}\) tuning range.
EC QCL linewidth = 25 MHz
Blue = QCL locked
Linewidth = 3 MHz

Error signal of QCL:
Most noise arises from High bandwidth temperature control feedback loop
CO$_2$ sub-Doppler spectroscopy
Sub-Doppler spectroscopy with EC QCL

Frequency accuracy: 10 - 100 kHz

Scan of CO$_2$ absorption features
Summary

- **Low noise OPO combs** pumped by high power Tm fiber combs
- OPOs are ready for **prime-time** in molecular spectroscopy
- Ultimate aim is to be better than **mass spectrometry**
- **QCLs** locked to fiber combs offer useful tools for precise line-shape analysis
- **Collisional effects** in multiple gas systems can now be studied
IMRA Prototypes

dual Tm comb system

- cw fiber pump lasers
- Control & feedback

High energy Tm fs source

- Tm, Ho, Yb fiber amplifiers
- Mid-IR laser technology
  - frequency conversion