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Dual-Comb Spectroscopy

Molecular Spectroscopy

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- Measures absorption of atoms and molecules as function of wavelength
- Depends on energy level structure
- IR frequency cause molecule to absorb energy and vibrate "fingerprint region"
  - Unique to each molecule, allow identification
  - Range from 10 to 100THz

Frequency Combs in Molecular Spectroscopy

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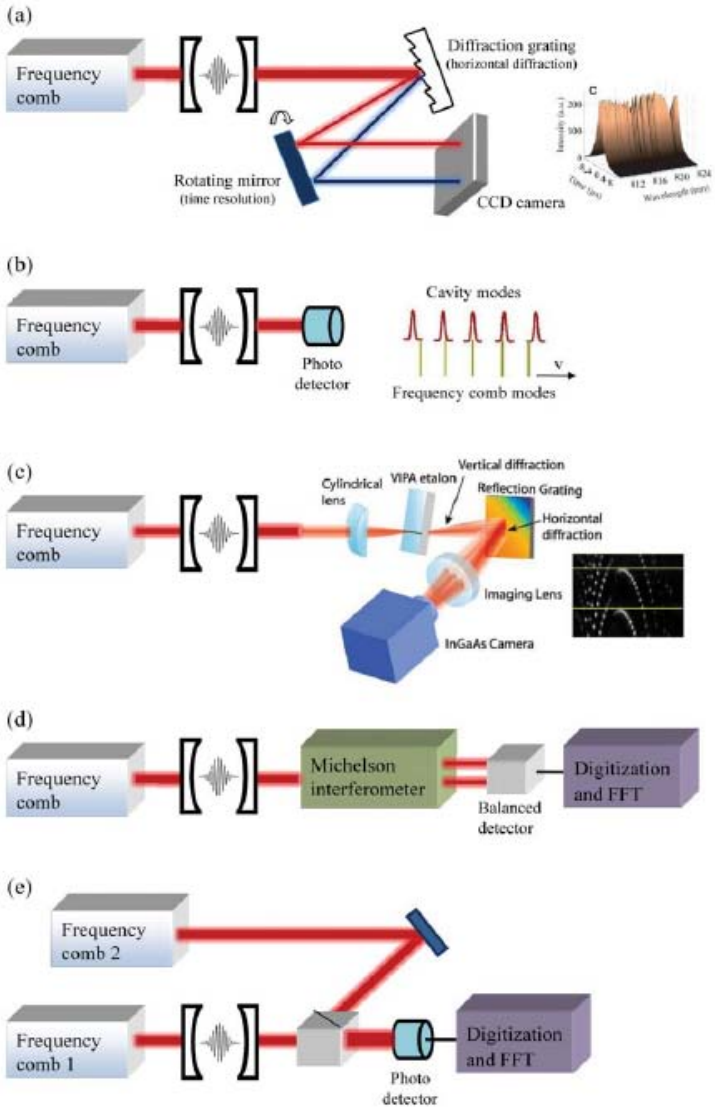
- Traditional Fourier transform spectroscopy
  - Broad band light source needed, long acquisition times
- Stabilized femtosecond lasers in the 90's revolutionized spectroscopy
  - Broad Bandwidth, Fine Resolution, Discrete sampling
  - Ideal for identifying absorption signatures of atoms and molecules, detecting trace gases
- Direct-Comb spectroscopy
- Dual Comb Spectroscopy – comparable to FT spectrometer with no moving parts, fast acquisition times



A. Foltynowicz… J.Ye, "Optical frequency comb spectroscopy," *RSC Faraday Discuss* **150**, 23-31 (2011)

Frequency Comb

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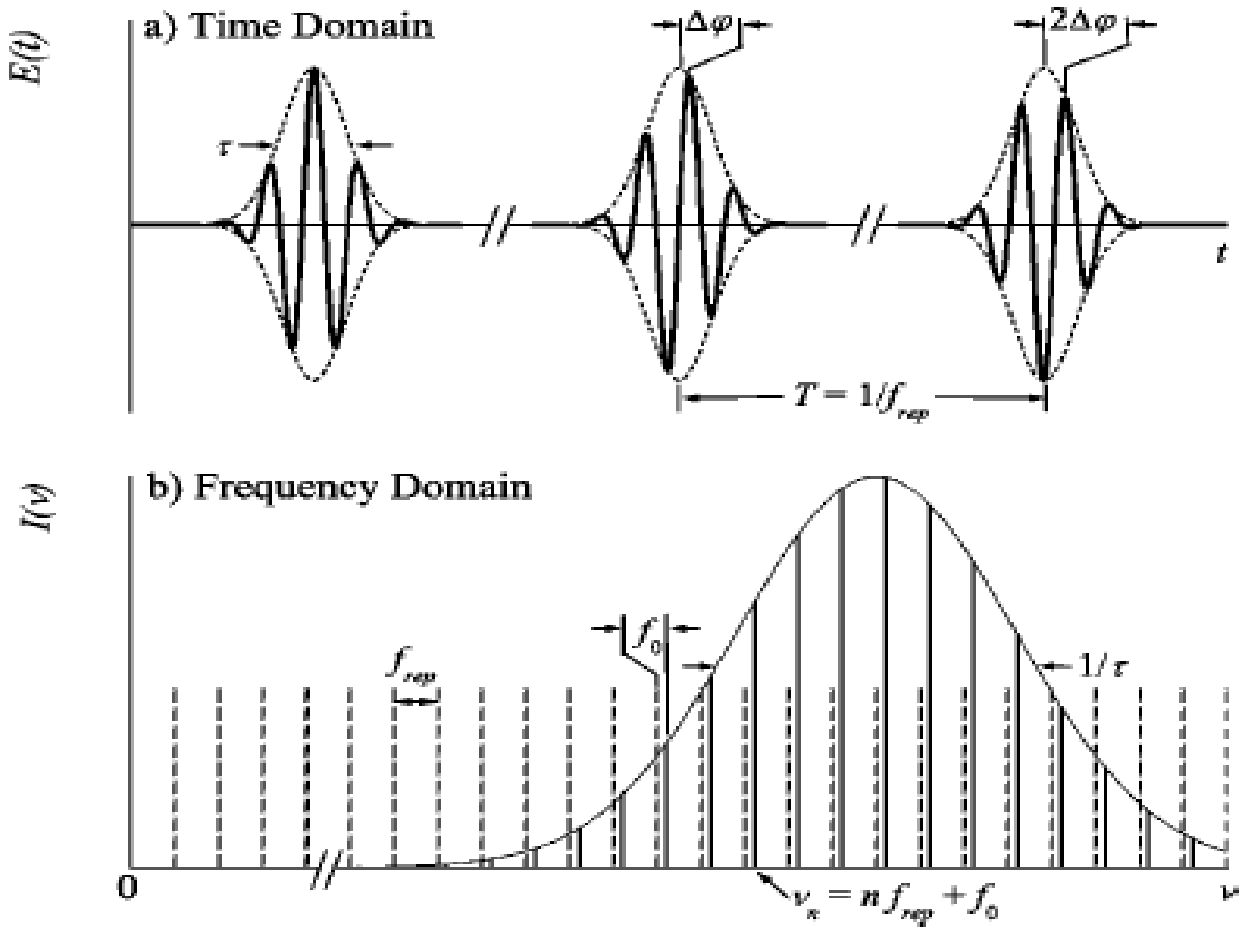

$$f_{n1} = f_{01} + nf_{rep1}$$

S. T. Cundiff, Jun Ye, "Colloquium: Femtosecond optical frequency combs," *Review of modern physics* **75,** (2003)

Basic Setup

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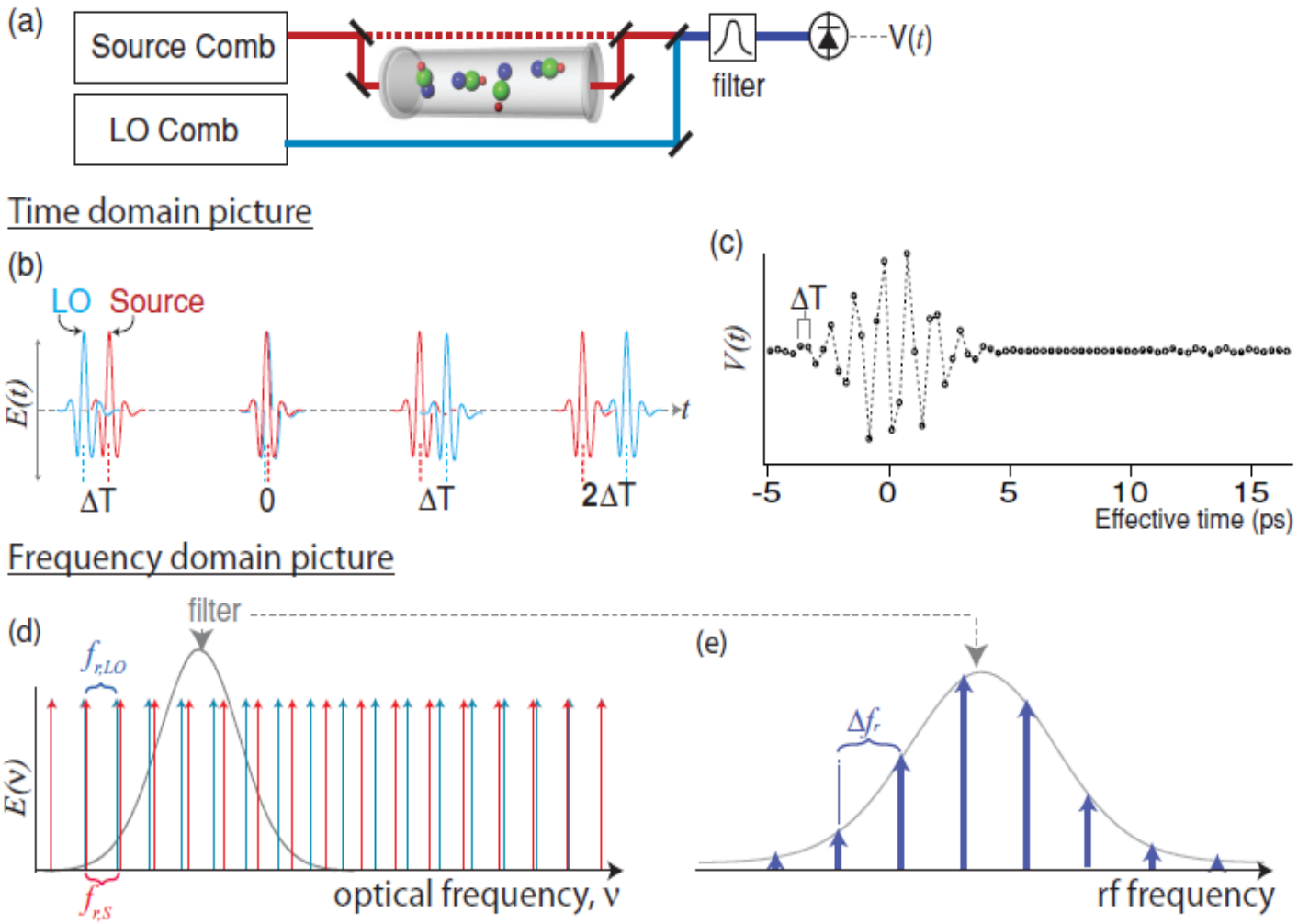

I. Coddington, W. C. Swann, and N. R. Newbury, " Coherent dual-comb spectroscopy at high signal-to-noise ratio," *Physical Review A* **82,** 043817 (2010)

Heterodyne detection

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- In order to have one-to-one mapping between RF and optical $\frac{\Delta v_{comb}}{f_r} < \left(\frac{f_r}{2\Delta f_r}\right)$
- FC1 $f_{n1} = f_{01} + nf_{rep1}$
- FC2 $f_{n2} = f_{02} + nf_{rep2}$
- Minimum acquisition time required $\frac{1}{\Delta f_r}$
- Beat Signal

$$I(t) = \sum_n A_n \cos\left[\left(f_{01} - f_{o2} + n\left(\left(f_{rep1} - f_{rep2}\right)\right)t\right]\right.$$

Stabilizing the comb

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- Frequency comb by itself has no inherent accuracy
  - Free running combs will drift
  - Needs to be monitored against some pair of absolute references.
  - With no reference you must derive results from known absorption line.

(Usually several orders of Magnitude worse then with Stabilized combs)

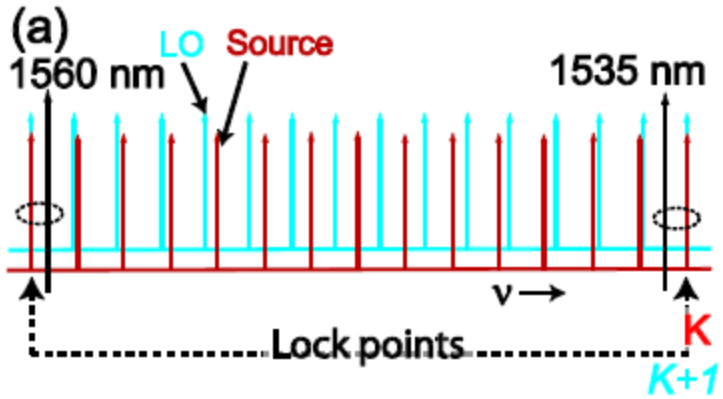

IMAGE: N. R. Newbury, " Coherent dual-comb spectroscopy at high signal-to-noise ratio," *Physical Review A* **82,** 043817 (2010)

Experimental Results

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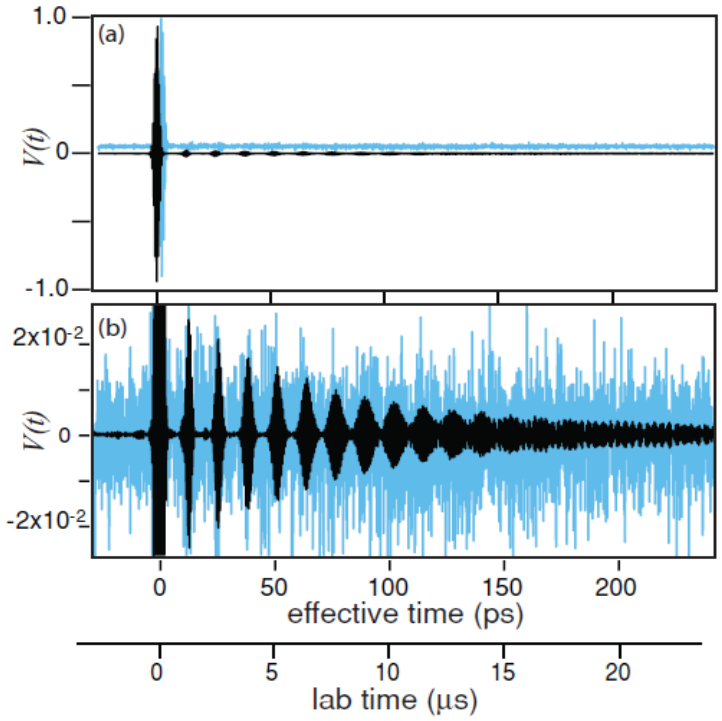

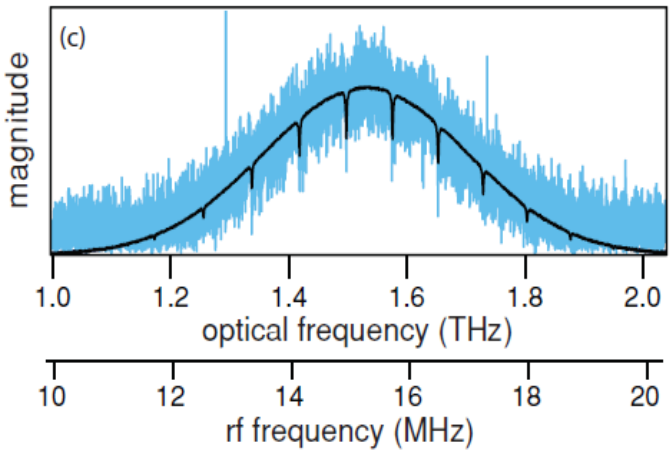

Sample: hydrogen cyanide (HCN)
around 1539nm
Filter set bandwidth at 350GHz
Acquisition time 1ms

$$\frac{1}{\Delta f_r}$$

I. Coddington, W. C. Swann, and N. R. Newbury, " Coherent dual-comb spectroscopy at high signal-to-noise ratio," *Physical Review A* **82,** 043817 (2010)

Cavity-enhanced dual-comb spectroscopy

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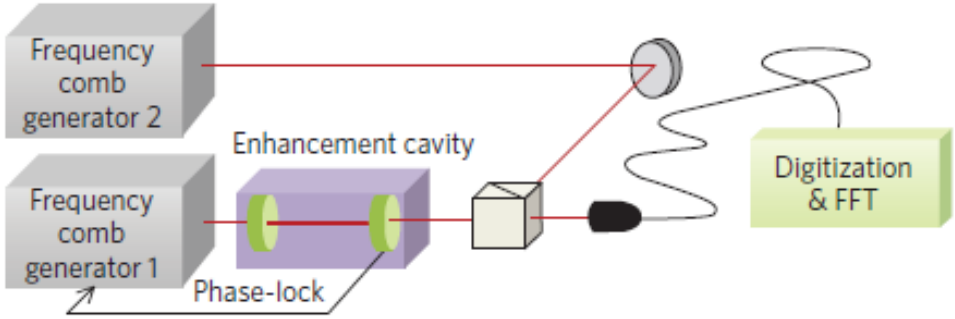
- Used in non-intrusive trace-gas sensing
  - Traditionally done with narrow bandwidth lasers
  - Recently FC's have been coupled to high-finesse cavity's but were detected with CCD arrays
    - 15nm spectral span, 25 GHz – 800 MHz resolution
    - $\alpha = 6.3(10^{-7})cm^{-1}\ to\ 8(10^{-10})cm^{-1}$
    - ms – tens of seconds for acquisition time
- Limitation of CCD arrays
  - Finer resolution meant changing comb parameters = longer acquisition times
  - Large detector arrays not available in mid-infrared molecular fingerprint region



- Dual Combs
  - 120nm spectral range, 5 GHz resolution
  - 42 us acquisition time
  - Sensitivity too low for trace gas detection
- Combining Dual-Comb with High Finesse cavity
  - Faster acquisition time (microseconds)
  - 20nm spectral span, 4.5GHz resolution

..Theodor W. Hansch, and Nathalie Picque, "Cavity-enhanced dual-comb spectroscopy," *Nature Photonics* **4**, 55-57 (2010)

Time Resolved Applications

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- The inteferogram periodicity

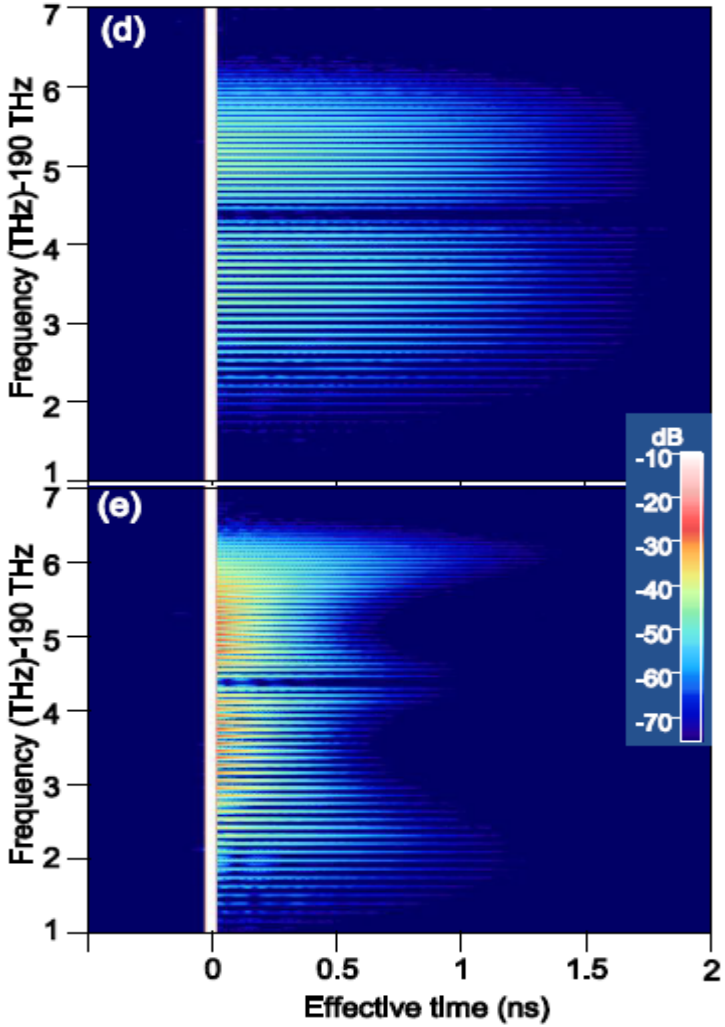
$$\frac{1}{f_{rep1} - f_{rep2}} = \frac{1}{\Delta f_r}$$

- Real-time monitoring of dynamic events
- Sonogram
  - 19GHz Frequency resolution and 52ps time resolution

  1st image shows lower pressure cell
  2nd image shows high pressure cell

  - At the lower pressure the decay is driven by Doppler rephrasing

    High pressure collisions accelerate the delay time



I. Coddington, W. C. Swann, and N. R. Newbury, " Coherent dual-comb spectroscopy at high signal-to-noise ratio," *Physical Review A* **82**, 043817 (2010)

Advantages

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- High Frequency Accuracy
  - 2-8 order of magnitude improvement in accuracy over state–of-the art FTS
  - High frequency resolution
    - At typical repetition rates can have 50-100MHz resolution
    - Traditional FTS hard to accomplish due to long travel distances in interferometer.
  - Acquisition rate very fast (only limited by Nyquist)
    - No moving parts

Limitations

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- Combs do not currently cover all parts of the spectrum that can be covered by traditional FTS
- Rely on highly stabilized combs

Questions?

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http://www.fseta.com/Uploads/Pages/Images/killing%20question.jpg