Optical frequency combs for fundamental metrology and gas spectroscopy

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Justervesenet - Norwegian Metrology Service
Outline

• Overview of Justervesenet – Norwegian Metrology Service

• National laboratory

• Time/frequency: central role in fundamental metrology

• Microwave vs optical atomic clocks

• Femtosecond frequency combs

• Direct frequency comb spectroscopy: Using a million stable laser lines
13 You shall not have in your bag differing weights, a large and a small.
14 You shall not have in your house differing measures, a large and a small.
15 You shall have a full and just weight; you shall have a full and just measure, that your days may be prolonged in the land which the LORD your God gives you.
16 For everyone who does these things, everyone who acts unjustly is an abomination to the LORD your God.

Deuteronomy 25

13 Du skal ikke ha to slags vektlodd i pungen din, noen store og noen små.
14 Du skal heller ikke ha to efa-mål i huset ditt, et stort og et lite.
15 Hele og rette vektlodd skal du ha og et helt og rett efa-mål. Da skal du få leve lenge i det landet Herren din Gud gir deg.
16 Herren din Gud avskyr enhver som gjør urett på denne måten.

5. Mosebok, kapittel 25

+ Metre convention (1875) + SI units
Justervesenet: Organization

**National laboratory**
Hans Arne Frøystein
(21 staff members)
- Electrometry, mass, fluid, time
  - Henning Kolbjørnsen
- Radiometry, temperature, length
  - Reidun A. Bergerud

14 MSc/cand.sc.
2 PhD/dr. ing
3 engineers

**Legal metrology**
Knut Lindløv
(45 staff members)
- Oslo verification office (at Kjeller)
- Stavanger verification office
- Bergen verification office
- Trondheim verification office
- Tromsø verification office

**Administration**
Anne Elisabeth Thorshov
(14 staff members)

**dir. general**
Ellen Stokstad
National Laboratory

Tasks:

Maintain national measurement standards for SI-units, ensure their traceability and trust

Calibrate standards and instruments for customers

Provide knowledge transfer in metrology-related issues (courses, consultancy, assessments etc.)

R&D
National Laboratory: Facilities

35 specially designed laboratories for accurate measurements
Excellent (and expensive – NOK 10 million/yr) environment control

Temperature ± 0.1 °C
Humidity ± 2.5 % RH
Vibration damping
RF - shielding

50 ton suspended concrete sub-floors
Expensive air conditioning
Quantum Hall Effect resistance
14 Tesla, 300 mK

Josephson effect DC voltage standard

Kilogram prototype no 36
### Quantity Realisation at Justervesenent

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Measurand</th>
<th>Realisation at Justervesenent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Length*</td>
<td>meter</td>
<td>Iodine-stabilized HeNe laser *</td>
</tr>
<tr>
<td>Time interval*</td>
<td>second</td>
<td>Cesium-clock *</td>
</tr>
<tr>
<td>Electrical DC voltage*</td>
<td>volt</td>
<td>Josephson-effect *</td>
</tr>
<tr>
<td></td>
<td></td>
<td>[ V_j = \frac{h}{2e} \bullet n \bullet f ]</td>
</tr>
<tr>
<td>Electrical DC resistance</td>
<td>ohm</td>
<td>Quantum Hall Effect</td>
</tr>
<tr>
<td></td>
<td></td>
<td>[ R = \frac{h}{e^2} = 25812.807449(86)\Omega ]</td>
</tr>
<tr>
<td>Mass</td>
<td>kilogram</td>
<td>National kilogram prototype</td>
</tr>
<tr>
<td>Temperature</td>
<td>Kelvin</td>
<td>Fixed Points</td>
</tr>
<tr>
<td>Luminous intensity</td>
<td>candela</td>
<td>Spectral response of detectors</td>
</tr>
<tr>
<td>Volume</td>
<td>litre or m³</td>
<td>Gravimetric and density measurements</td>
</tr>
</tbody>
</table>

* Directly related to a stable frequency reference

Paradox (?): The least tangible physical quantity is the most precisely defined and plays a key role in the physical realisation of other units.
Microwave vs optical atomic clocks

Stability of current Cs clocks limited by the linewidth (100 Hz) relative to the absolute frequency of the 133Cs hyperfine transition (9 GHz). Ultimate relative uncertainty $10^{-15}$.

Sharp optical transitions (linewidth sub 1 Hz) relative to optical frequencies (400 THz) may ultimately enable optical oscillators with a relative uncertainty $10^{-19}$.

How can stable oscillators at optical frequencies be used as a clock?

- Problem: There are no frequency counters in the THz range
- Solution: Optical frequency combs
Femtosecond optical frequency combs

- Tool for measuring optical frequencies
- Optical frequency ‘ruler’
- Produced by lasers emitting a regularly spaced train of ultrashort pulses
- Or a laser with a million stable lines for spectroscopy
- Nobel prize in physics 2005 awarded John L. Hall and Theodor W. Hänsch "for their contributions to the development of laser-based precision spectroscopy, including the optical frequency comb technique"
Mode locked laser

- Many modes are amplified simultaneously
- When the phase between different modes is locked, the resulting emission consists of short pulses
Time domain $\rightarrow$ frequency domain

- Pulse repetition rate $f_r$:
  - Comb mode spacing

- Spectral width
  - Inversely proportional to the pulse width $\tau$
Frequency domain

Optical frequencies (100s THz) defined by two RF frequencies and a large integer

\[ f_n = n f_r + f_{\text{offset}} \]
Offset frequency

- Offset-frequency $f_{\text{offset}}$ is the phase shift between pulses
- Due to different phase- and group dispersion
Stabilizing frequency combs

• The pulse repetition rate is measured directly with a fast photodiode and locked to a stable frequency reference (e.g. Cs atomic clock) by adjusting the laser cavity length.

• The offset frequency is measured by beating one tooth of the comb with its 2nd harmonic. $f_{\text{offset}}$ is locked to a stable frequency reference by e.g. modulating the laser pumping power.

• The absolute frequency stability of combs is currently limited by the stability of Cs clocks. Relative stability between different combs demonstrated at $10^{-19}$ level.

Measuring the offset frequency of an octave spanning frequency comb
Measuring absolute optical frequencies

• Measure the beat frequency between the stabilized comb and a stable laser

• Measure the pulse repetition rate directly

• $n$ is determined by measuring the approximate frequency with a less accurate wave meter

• Offset frequency measured by self-referencing

$$f_n = nf_r + f_{\text{offset}}$$
Direct frequency comb spectroscopy

How to use a million stable laser lines for spectroscopy?

- As a brilliant (super-)continuum source for FT spectroscopy
- Direct spectroscopy with optical heterodyne detection
- Cavity filtered and
- Cavity enhanced spectroscopy
Commercial FT spectrometer
Non-stabilized frequency comb used as a high brilliance broadband source
SNR improved a factor 17 over tungsten source
Two stabilized frequency combs required

Absorption magnitude and phase recovered from interference between local oscillator and sample probe laser
FIG. 3 (color online). (a) Measured transmission (black) and phase (gray) spectrum for HCN spanning 15.5 THz of optical spectrum (from 1495 to 1620 nm). There are ~155,000 individual points each corresponding to a single heterodyne beat, such as those shown in Fig. 2. The power per comb tooth varied from 2 nW in the center of the spectrum to 20 pW on the edges of the spectrum. (b) Expanded view covering the 4.5 THz containing the HCN absorption lines. (c) Further expanded view covering 0.3 THz. The measured phase agrees well with the calculated phase (dotted blue line, offset by 0.1 rad) from the Kramers-Kronig transformation of the absorption data. The standard deviation of the phase measurement is 1.6 mrad. The measured absorption agrees well with previous published data [22] (dashed blue line, offset by 0.2).
Fabry-Perot cavity acts as a filter selecting a subset of comb modes. Stabilized comb required for controlled coupling of modes into the cavity. Transmitted light spatially dispersed in two stages VIPA + grating.
Scanning the repetition frequency enables extremely fine resolution.
Cavity-enhanced optical frequency comb spectroscopy: application to human breath analysis

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Sensitivity enhancement with the gas in a high finesse cavity (i.e. many passes)
Coupling of comb modes to the cavity requires tight stabilization

Michael Thorpe’s PhD thesis available at: http://www.colorado.edu/YeLabs
Fig. 5. A breath spectrum between 1.622 μm and 1.638 μm. Several windows in this region contain spectroscopic features of three isotopes of CO₂ with nearly equal absorption strengths. (a) and (b) Two zoomed-in spectral windows where line positions and intensities of relevant CO₂ transitions are shown. Besides CO₂ peaks, strong absorption features of H₂O and CH₄ are detected in (b). (c) The entire range over which this condition exists.

Fig. 6. (a) The CO₂ and CO absorption spectra of student 1 (smoker) and student 2 (non-smoker) in the 1.564 μm spectral region, along with line intensities from the HITRAN spectral database. The smoker’s obvious increase in CO concentration is clearly detected. (b) The breath concentration of CO₂ in parts per thousand (ppt) and CO in parts per million (ppm) as a function of time during which the test subject holds their breath prior to exhaling into the sample bag.
Summary

- Stabilized frequency combs will become the workhorse of future precision metrology

- A million stable laser lines offer new opportunities for sensitive spectroscopy

- The cost per laser line is very small (NOK 1)!