# Viewpoint

## Ultrasensitive Absorption Spectroscopy

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*Absorption spectroscopy limited only by quantum noise has been demonstrated over a broad frequency range using laser frequency combs.*

Subject Areas: **Atomic and Molecular Physics, Optics**

**A Viewpoint on:**
**Quantum-Noise-Limited Optical Frequency Comb Spectroscopy**
Aleksandra Foltynowicz, Ticijana Ban, Piotr Masłowski, Florian Adler, and Jun Ye
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Absorption spectra have been widely used for decades to study the structure and dynamics of atoms and molecules. In fundamental research they permit the extraction of thermodynamic and kinetic properties, while in the applied domain they are employed as analytical tools to detect the presence of a particular compound or its concentration in a sample. Nowadays, optical absorption spectroscopy is a powerful tool used for many applications in physics, chemistry, biology, and medical sciences.

A wide range of experimental approaches to measure absorption spectra have been developed and successfully employed for detecting molecules. The light source, sample arrangement, and detection technique vary significantly depending on the frequency range and the purpose of the experiment. All the established methods demonstrate that there is no perfect spectroscopic method that can satisfy the needs of all applications, and a compromise is needed between spectral coverage, resolution, sensitivity, and acquisition time. However, increasingly demanding scientific goals and new requirements in molecular physics require new methods that combine fast acquisition times, high spectral resolution, broad spectral coverage, high detection sensitivity, and accurate optical frequency measurements. In this context, in a paper appearing in *Physical Review Letters*, Aleksandra Foltynowicz and colleagues, from the National Institute of Standards and Technology and the University of Colorado in Boulder have implemented advanced spectroscopic instrumentation over a broad frequency range with a laser frequency comb that is limited only by quantum noise [1].

Recently developed, laser frequency combs are attractive light sources for absorption spectroscopy [2, 3] and have been used for proof-of-principle studies in the infrared region [4, 5]. Equivalent to a million monochromatic lasers emitting at equidistant frequencies spanning an octave, they combine the advantages of broad spectral coverage and high spectral brightness. Moreover, frequency combs enable precise measurements of the electromagnetic spectrum, as it is now straightforward to determine the absolute frequencies of all comb lines. By wisely coupling the frequency comb source with a high-finesse cavity and a Fourier spectrometer, Foltynowicz and co-workers have achieved the highest sensitivity per spectral element for a comb-based technique.

In spectroscopy based on optical frequency combs, enhancement of the sensitivity can be easily attained by increasing the interaction length between the light source and the sample. A straightforward way is to use a multipass cell with a long optical path [6], sufficient enough for simultaneous acquisition of the broadband spectra of many molecular species at concentrations in the parts-per-billion range. The second possibility is to use a high-finesse cavity. The finesse of an optical cavity is defined as the spectral range divided by the bandwidth of its resonances. It is therefore natural that a high-finesse cavity associated with a wide spectral range and narrow resonances is suitable for optical spectrum analysis. The use of a Fabry-Pérot cavity with a frequency comb has already been proposed [7, 8], and when the multipass cell is replaced with a high-finesse cavity, the sensitivity is expected to improve by few orders of magnitude, providing a parts-per-trillion level detection limit. However, due to the cavity dispersion mainly introduced by the mirrors, the cavity modes are not uniformly spaced, producing a cavity resonant frequency structure that the comb cannot match over its full wavelength range. Therefore it is still challenging to benefit from the full spectral bandwidth offered by the frequency comb laser.

Foltynowicz *et al.* address this problem by implementing a two-point locking scheme. It consists of locking the frequency comb at two different wavelengths to maximize the transmission through the cavity and to cou-

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ple the widest spectral range possible. In this way, a 50-nanometer-wide wavelength region (at a wavelength of 1530 nanometers) is simultaneously coupled into the cavity. The experience of the group in locking systems allowed the frequency comb to be tightly locked to the cavity in order to ensure constant transmission and avoid the frequency-to-amplitude noise conversion that could potentially compromise the sensitivity.

In the recent years, motivated by the desire to take advantage of all the spectral properties of the frequency comb as a light source for high-resolution absorption spectroscopy, several detection schemes have been proposed. One of them consists of using dispersive elements with a multidetector array [2, 9]. With this approach, the acquisition time can be relatively short, of the order of a few milliseconds, but a tradeoff is made by diminishing either the spectral coverage or the spectral resolution, limited to a few nanometers or a few hundred of megahertz, respectively. Another possibility is to use Fourier transform spectrometers. This method offers no limitation concerning spectral coverage and resolution and can be achieved either with a mechanical spectrometer based on a Michelson interferometer [5, 6] or with dual frequency comb spectroscopy that offers unprecedented qualities in terms of spectral resolution and recording time [10, 11].

By using a Fourier spectrometer based on a Michelson interferometer, Foltynowicz and co-workers have simultaneously recorded all the spectral elements transmitted by the high-finesse cavity; the latter remains the only spectral limitation of the method. A schematic of the setup is shown in Fig. 1. The demonstrated extreme sensitivity is achieved not only by using a high-finesse cavity but also an auto balancing detector that was built on the basis of a Hobbs design [12]. The output of the interferometer is made of two beams containing the modulated interferogram in opposite phases. As the two beams are derived from the same light source, laser noise and noise generated by the high-finesse cavity will appear coherently in both the beams. The balanced detection subtracts the signal for the two optical beams, which is useful for two reasons. First, it removes part of the interferogram that is not modulated by the path difference, thereby increasing the dynamic range of the measurements. Second, it cancels the coherent noise, thus increasing the detection sensitivity. This discussed detection system also avoids nonlinear effects, which can appear due to the high intensities of the femtosecond pulses. Combining all these elements, the method demonstrates its suitability, by recording the spectrum of acetylene, for high-resolution broadband spectroscopy with high sensitivity and stable operation over hours.

In conclusion, frequency combs are triggering new approaches for broadband spectroscopy. Foltynowicz and co-workers have successfully combined the frequency comb with advanced techniques known from absorption spectroscopy and reached, for the first time, quantum-noise limited detection over a broad spectral domain at high spectral resolution. The locking system of the laser to the cavity and the Fourier spectrometer allowed maintaining a shot-noise-limited system over hours, making the system ready to use for numerous applications. Moreover, the system can be easily implemented at any wavelength, and in particular in the mid-infrared region for ultrasensitive multispecies detection, as needed for breath analysis or detection of hazardous gases.

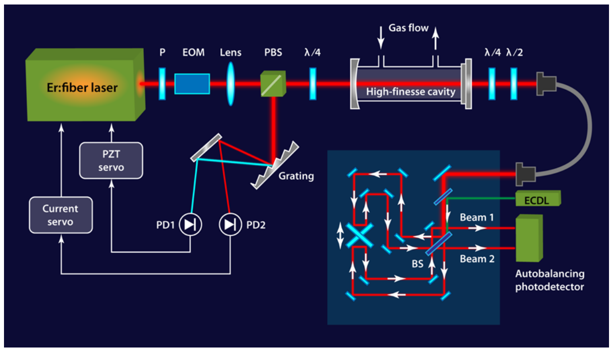


FIG. 1: Schematic of the setup for ultrasensitive detection of gas species by measuring their absorption spectra with a laser frequency comb source. An Er:fiber femtosecond laser is locked to a high-finesse optical cavity containing the gas sample. An electro-optic modulator (EOM) is used to phase modulate the comb light at 14 megahertz, and the cavity reflected light is dispersed by a reflection grating and imaged on two photo detectors (PD1 and PD2) in order to create error signals at two different wavelengths. The feedback is sent to a laser current controller and to a PZT (piezoelectric transducer) inside the laser cavity. The cavity transmitted light is coupled through a polarization-maintaining fiber into a fast-scanning Fourier transform spectrometer. The two outputs of the interferometer (beams 1 and 2) are incident on two photodiodes of the autobalancing photodetector. The beam of a continuous-wave 780-nanometer external cavity diode laser (ECDL), used for frequency calibration, is propagating parallel to the frequency comb beam and incident on a separate detector (not shown). P = polarizer, PBS = polarizing beam splitter cube, $\lambda/4$ = quarter-wave plate, $\lambda/2$ = half-wave plate. (APS/Alan Stonebraker)

## References

[1] A. Foltynowicz, T. Ban, P. Masłowski, F. Adler, and J. Ye, *Phys. Rev. Lett.* **107**, 233002 (2011).
[2] M. J. Thorpe *et al.*, *Science* **311**, 1595 (2006).
[3] S. A. Diddams, L. Hollberg, and V. Mbele, *Nature* **445**, 627 (2007).
[4] E. Sorokin *et al.*, *Opt. Express* **15**, 16540 (2007).
[5] F. Adler *et al.*, *Opt. Express* **18**, 21861 (2010).
[6] J. Mandon, G. Guelachvili, and N. Picque, *Nature Photon.* **3**, 99 (2009).
[7] M. J. Thorpe and J. Ye, *Appl. Phys. B* **91**, 397 (2008).
[8] F. Adler *et al.*, *Annu. Rev. Anal. Chem.* **3**, 175 (2010).
[9] C. Gohle *et al.*, *Phys. Rev. Lett.* **99**, 263902 (2007).
[10] I. Coddington, W. C. Swann, and N. R. Newbury, *Phys. Rev. Lett.* **100**, 013902 (2008).
[11] B. Bernhardt *et al.*, *Nature Photon.* **4**, 55 (2010).

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[12] P. C. D. Hobbs, *Appl. Optics* **36**, 903 (1997).

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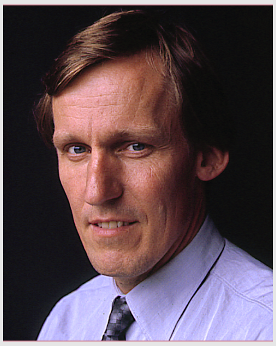
Julien Mandon is a postdoctoral fellow at the Radboud University, Nijmegen (the Netherlands). He completed his Ph.D. in physics in 2009 at the University Paris Sud, Orsay (France), where he used frequency combs for infrared spectroscopy and proposed, for the first time, its use in combination with Fourier spectroscopy. Financed by an EU Marie Curie fellowship, his recent research involves the use of quantum cascade lasers with innovative infrared spectroscopic schemes for the detection of trace gas emitted by various biological samples.

### Frans Harren



Frans J. M. Harren is Associate Professor at Radboud University, Nijmegen (the Netherlands). His research focuses on the reliable state-of-the-art sensing of minute quantities of trace gases in complicated gas mixtures, online, noninvasive, with high selectivity and detection speed. For this, photoacoustic frequency modulation, and cavity-enhanced spectroscopy are combined with quantum cascade lasers, optical parametric oscillators, and frequency comb lasers. These methods are applied within biology, agro technology, and medical sciences.

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