Frequency comb spectroscopy with coherent optical sampling

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Abstract: A stabilized frequency comb provides a broadband array of highly resolved comb lines. Using a multiheterodyne technique, we measure the amplitude and phase of every comb line, allowing for massively parallel, high-resolution spectroscopy.

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1. Introduction

One can think of a stabilized frequency comb as a train of femtosecond pulses that are effectively the sum of a hundred thousand or more single frequency lasers (often referred to as comb teeth) whose frequencies are spread evenly across broad portions of the optical spectrum. This melding offers an alluring combination: the frequency precision, stability and resolution of continuous wave lasers coupled with the enormous bandwidth and time resolution of femtosecond pulses. As such, these combs can be used for massively parallel spectroscopy in which 150,000 comb teeth spanning 120 nanometers are used to record the absorption and phase spectra of a gas sample [1]. The fine time resolution of the femtosecond pulses also allows us to observe very fast temporal responses such as free induction decay or temporally delayed, forward scattered light from the gas.

The stabilized femtosecond frequency comb was originally developed as a powerful tool for frequency metrology [2, 3] providing stable and well known frequencies over much of the optical spectrum (see figure 1). The challenge for spectroscopy is to simultaneously resolve between a hundred thousand and a million individual comb lines. Expanding on earlier work by Kielman *et. al.*[1, 4-9] we demonstrate that this separation can be done by heterodyning the frequency comb with a second comb of slightly different frequency spacing. The result of this heterodyning is a radio frequency (rf) comb where each tooth corresponds to the heterodyne beat between a single optical tooth from each comb.

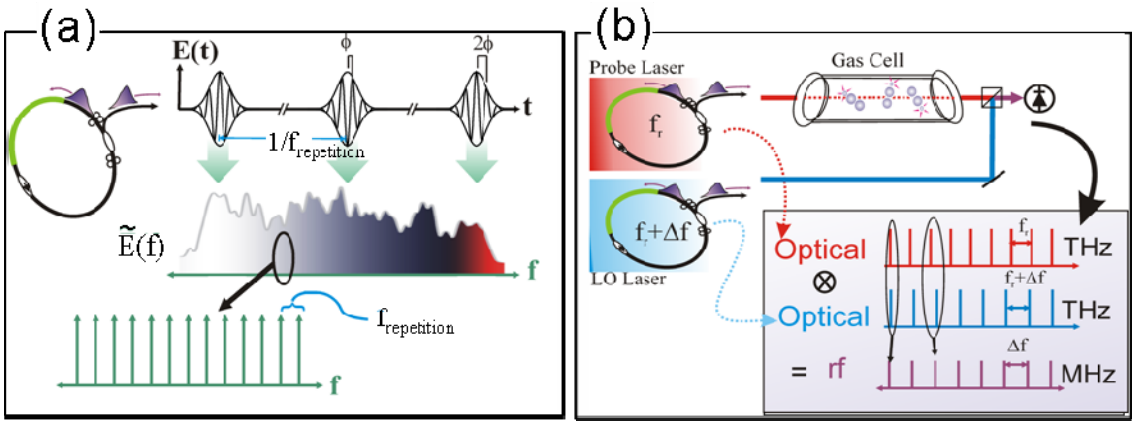

Figure 1: (a) A femtosecond fiber laser emits a train of pulses in time at a repetition frequency $f_{repetition}$. The spectrum of each pulse extends over 125 nm and can be further broadened in nonlinear optical fiber, but retains the "comb" structure. (b) Basic concept behind multiheterodyne spectroscopy. The two combs are slightly offset in repetition rate. As a result, their heterodyne signal leads to an rf comb, with a one-to-one mapping between the optical and rf comb teeth.

The advantage of this multiheterodyne scheme is at least four-fold. First, the mapping of the optical comb into the rf allows for the straightforward retrieval of each comb line by use of a single photodiode and a fast digitizer. Secondly, the mapping also allows for near perfect knowledge of absolute frequency. Thirdly, the heterodyne signal maps the full electric field, meaning that both optical amplitude and optical phase are simultaneously retrieved. Finally, the use of heterodyne detection allows for the detection of very weak signals and the rejection of much technical noise. We were able to retrieve data

with as little as twenty picowatts per comb tooth, and with an acquisition time of 1 second per nanometer of spectrum.

2. Experiment

For this experiment we use a pair of erbium fiber-based femtosecond frequency combs. Each comb has a bandwidth of roughly 125 nm centered around 1550 nm. The repetition rates (tooth spacing) of the two combs, 100,016 kHz and 100,017 kHz, cause the slight frequency mismatch necessary for our rf mapping detection scheme.

Figure 1(b) shows the experimental configuration for using two frequency combs to measure the absorption response of a hydrogen cyanide (HCN) gas sample. The probe laser is used to sample the gas and the second laser acts as a local oscillator (LO) and records changes in the probe spectrum due to the gas. Here we emphasize the spectroscopic applications, but in principle one could use this system to query the optical response of a number of systems such as telecom components or nonlinear materials. In addition to the beam path shown in Figure 1(b) we also incorporate a dummy path without a gas cell. This path allows us measure the background amplitude and phase profile of the combs, which is later subtracted out to isolate the spectrum of the HCN gas.

Because we simultaneously resolve multiple heterodyne beats, it is critical that we have a high degree of stability between the two frequency combs. We achieve this stability by phase locking each comb to the same pair of single frequency lasers located at 1535 nm and 1550 nm. This is sufficient to stabilize the two degrees of freedom in a frequency comb and allows us to achieve a linewidth of less than one hertz between the two combs. More information on this technique can be found in Refs [1, 10, 11].

Figure 2(a) shows the resulting phase and transmittance spectrum of our HCN sample. Across the figure there are 50,000 data points, each corresponding to the phase shift and absorption experienced by a single comb tooth.

It is also interesting to consider the time domain picture, where the approach is really one of coherent linear optical sampling.[12]. Recall that we have a slight mismatch in repetition rates for our two lasers, the result being that the pulses from the LO laser slowly march through, and sample, the probe pulses. The 100 femtosecond nature of the pulses acts as a very fine sampling gate and in effect one can think of this system as a highly resolved cross correlation in time with the LO laser sampling the probe laser. In truth the rawest form of our data is not the frequency picture shown above but rather a very high resolution time image of the pulse after it has traveled through the gas. Figure 2(b) shows a time trace of such a pulse that is followed by a tail of smaller pulses. This tail was first observed by Brewer and Shoemaker[13] and corresponds directly to the re-phasing and de-phasing of rotating molecules excited by the pulse, also referred to as free induction decay (FID).

While our results here are preliminary, it is interesting to note that this FID signal has a very specific frequency, phase and decay. Additionally, it exists on a dark background. This type of time-domain signature can only be inferred from standard absorption spectroscopy data. Here, however, it can be directly measured using the narrow time gating of a frequency comb system. This time-domain signature is

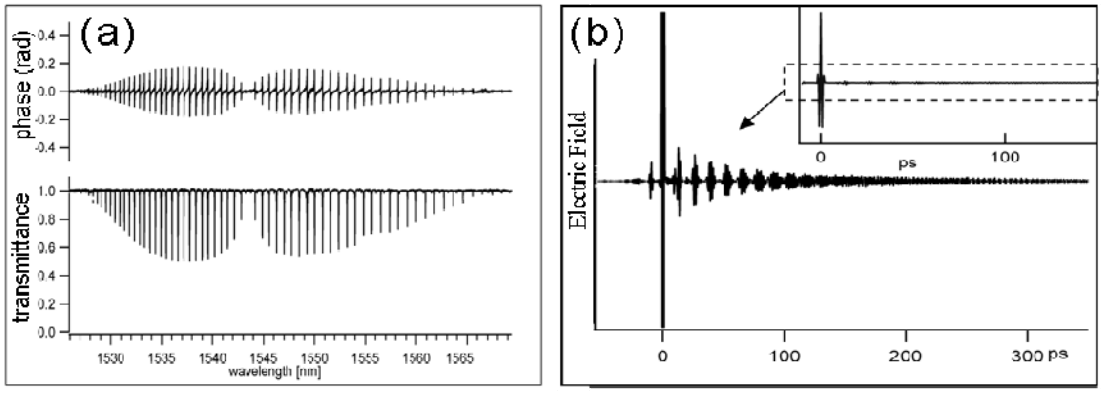

Figure 2(a) Measured phase and absorption spectrum for hydrogen cyanide (HCN). In total the measurement spanned from 1492 nm to 1618 nm or 155,000 individual frequency comb lines. For clarity only the data showing HCN absorption bands is shown. Data is in excellent agreement with previously published values [8]. (b) Temporal cross correlation in time between pulses from the two combs. The large spike is the original pulse and is followed by a free induction decay signal that corresponds to the rotation of excited dipoles. For clarity, only 10 nm of optical spectrum was used here, covering ~6 rovibrational lines.

interesting both from a fundamental point of view, and because it has potential as a very sensitive means of performing broadband gas detection. Because the signature is composed entirely of evenly spaced frequencies that originated with the frequency comb it could be combined with a cavity buildup scheme to greatly extend the absorption path length of the gas.[14]

In the time domain picture there also exist some interesting analogies to standard FTS systems.[15] One can think of the LO comb as a virtual delay line in that it interferes with the signal laser at a series of different delays. In an FTS system the coherence between the two delay line arms is guaranteed by the common source. The price of our system is that we must force coherence between our lasers through phase locks. In this comparison phase noise between the lasers is equivalent to mechanical jitter in the delay line. However it quickly becomes clear that one strength of our system is speed. The LO scans the entire 10 ns repetition period of the signal laser in $t=1/\Delta f$ (1 millisecond in our case). Thus our virtual delay line is equivalent to a system that can scan 1.5 meters at a rate of 1.5 km/s. Clearly such a delay line is beyond current technology.

3. Conclusion

Stabilized femtosecond fiber lasers produce broadband coherent light that can be exploited in a number of high-resolution measurements. Originally, these combs found their main application in optical frequency metrology, but they should also find many other applications in other areas of high-resolution measurement.

References

1. I. Coddington, W. C. Swann, and N. R. Newbury, "Coherent Multiheterodyne Spectroscopy Using Stabilized Optical Frequency Combs," Phys. Rev. Lett. 100, 103902-013906 (2008).
2. J. L. Hall, " Nobel Lecture: Defining and measuring optical frequencies," Rev. Mod. Phys. 78, 1279 (2006).
3. T. W. Hänsch, " Nobel Lecture: Passion for precision," Rev. Mod. Phys. 78, 1297 (2006).
4. S. Schiller, "Spectrometry with frequency combs," Opt. Lett. 27, 766-768 (2002).
5. T. Yasui, Y. Kabetani, E. Saneyoshi, S. Yokoyama, and T. Araki, "Terahertz frequency comb by multifrequency-heterodyning photoconductive detection for high-accuracy, high-resolution terahertz spectroscopy," Appl. Phys. Lett. 88, 241104-241101 - 241104-241103 (2006).
6. A. Schliesser, M. Brehm, F. Keilmann, and D. v. d. Weide, "Frequency-comb infrared spectrometer for rapid, remote chemical sensing," Opt. Express 13, 9029-9038 (2005).
7. P. Giaccari, J.-D. Deschenes, P. Saucier, J. Genest, and P. Tremblay, "Active Fourier-transform spectroscopy combining the direct RF beating of two fiber-based mode-locked lasers with a novel referencing method," Opt. Express 16, 4347-4365 (2008).
8. A. Schlatter, S. C. Zeller, R. Pashcotta, and U. Keller, "Simultaneous measurement of the phase nosie on all optical modes of a mode-locked laser," Appl. Phys. B 88, 385-391 (2007).
9. F. Keilmann, C. Gohle, and R. Holzwarth, "Time-domain and mid-infrared frequency-comb spectrometer," Opt. Lett. 29, 1542-1544 (2004).
10. W. C. Swann, J. J. McFerran, I. Coddington, N. R. Newbury, I. Hartl, M. E. Fermann, P. S. Westbrook, J. W. Nicholson, K. S. Feder, C. Langrock, and M. M. Fejer, "Fiber-laser frequency combs with sub-hertz relative linewidths," Opt. Lett. 31, 3046-3048 (2006).
11. N. R. Newbury, and W. C. Swann, "Low-noise fiber laser frequency combs," J. Opt. Soc. Am. B 24, 1756-1770 (2007).
12. C. Dorrer, D. C. Kilper, H. R. Stuart, G. Raybon, and M. G. Raymer, "Linear Optical Sampling," Photonics Technology Letters 15, 1746-1748 (2003).
13. R. G. Brewer, and R. L. Shoemaker, "Photon Echo and Optical Nutation in Molecules," Phys. Rev. Lett. 27, 631 (1971).
14. M. J. Thorpe, D. Hudson, K. D. Moll, J. Lasri, and J. Ye, "Cavity-ringdown molecular spectroscopy based on an optical frequency comb at 1.45-1.65 um," Opt. Lett. 32, 307-309 (2007).
15. J. Mandon, G. Guelachvili, N. Picque, F. Druon, and P. Georges, "Femtosecond laser Fourier transform absorption spectroscopy," Opt. Lett. 32, 1677-1679 (2007).