Time and Frequency-Domain Spectroscopy with Dual Frequency Combs

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Abstract: High-resolution spectroscopic measurements of the amplitude and phase spectra from a gas sample can be acquired by use of dual frequency combs. Here we discuss the corresponding gas signature in the time domain.

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One can think of a stabilized frequency comb as a train of femtosecond pulses that are effectively the sum of one hundred thousand or more single-frequency lasers (the comb teeth). This is an alluring combination: the frequency precision, stability and resolution of continuous-wave lasers coupled with the enormous bandwidth and time resolution of femtosecond pulses. We recently demonstrated that this system 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 hydrogen cyanide (HCN) gas sample.[1] Here, we point out that the fine time resolution of the femtosecond pulses effectively allows us to observe free induction decay or temporally delayed, forward scattered light from the gas. This light is interesting in that it has very specific frequency and decay signatures and exists on a potentially shot-noise limited background. This forward-scattered light allows for both frequency and temporal discrimination of gasses and in principle will allow for sensitive detection of trace gasses.

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 address between one-hundred thousand and a million individual comb lines. Recently there has been significant progress in resolving the individual comb teeth after they pass through a gas or other sample.[4, 5] We and others have taken the approach of heterodyning the frequency comb with a second comb of slightly different frequency spacing.[1, 6-11] For sufficiently narrow linewidth fiber combs,[12] the result of this heterodyne signal is a radio frequency (rf) comb where each tooth corresponds to the heterodyne beat between a single optical tooth from each comb.

The advantage of this scheme is at least four-fold. First, the mapping of optical combs 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 is really a mapping of 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.

It is interesting to consider the time-domain picture, where the approach is really one of coherent linear optical sampling.[13-15] Recall that we have a slight mismatch in repetition rates for our two lasers. One can think of this as the pulses from one laser slowly marching past the other in time. The short LO pulses then act as a very fine sampling gate, and in effect this system performs a highly resolved cross-correlation in time. In truth the rawest form of our data is not the frequency picture shown in Ref. [1], but rather a very high resolution time optical waveform sampling of the broadband laser pulses exiting the gas. Figures 1(b) shows a time traces of such a pulse, which is followed by a tail of smaller pulses. This tail was observed by Brewer [16] 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).

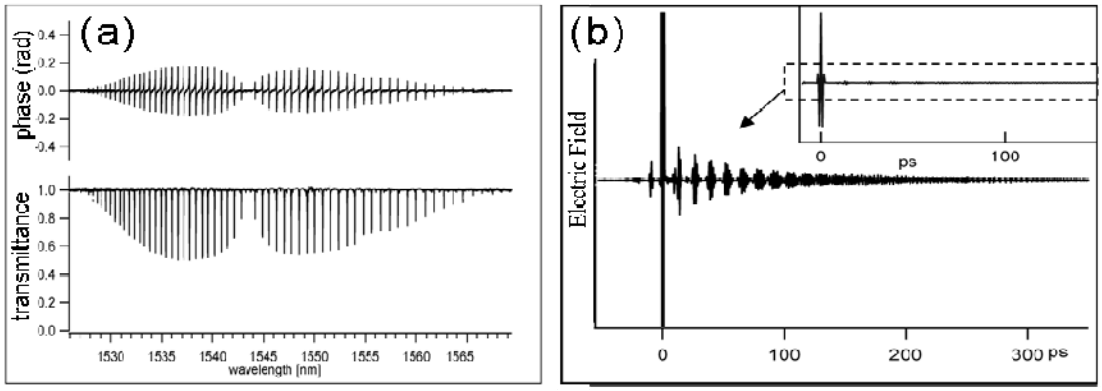

Figure 1(a) Measured phase and absorption spectrum for Hydrogen Cyanide (HCN). In total the measurement spanned from 1492 nm to 1618 nm or 155,000 individual frequency comb lines. For clarity, only the data showing HCN absorption bands are shown. Data are in excellent agreement with previously published values [8]. (b) Cross correlation in time between pulses from the two combs. The large spike at zero time 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.

While our results here are preliminary,the FID signal is clearly observed with its a very specific frequency, phase and decay time depending on the molecular gas species. (The pulse at earlier times in Fig 1b is on the actual laser output and not from the gas itself.) This type of time-domain signature can be inferred only from standard absorption spectroscopy data, but it can be directly measured in the dual-comb system. The time-domain signature is interesting both from a fundamental point of view, and because it has the potential to become a very sensitive means of measuring broadband gas detection, particularly if it is combined with a cavity buildup scheme to effectively increase the absorption path length of the gas.[17]

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