Performance of a Coherent Dual Frequency Comb Spectrometer

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Abstract: We discuss the performance of a coherent dual frequency comb spectrometer in terms of signal-to-noise ratio, resolution, and accuracy based on experimental data in the near and short-wave infrared centered at 1.5 and 3.4 μm.

OCIS codes: 120.0120 (Instrumentation, measurement, and metrology), 320.7090 (Ultrafast lasers)

1. Introduction

The broadband spectrum and coherent output of frequency combs has sparked much interest in using frequency combs directly for optical spectroscopy. One particular method that has been explored in recent years is coherent dual-comb spectroscopy, alternatively called multi-heterodyne or frequency-comb spectrometer [1-11]. This new method of comb spectroscopy has promise as a spectroscopic tool. Its performance falls somewhere between conventional Fourier transform spectroscopy (FTS) and cw laser heterodyne spectroscopy in terms of spectral coverage, SNR, frequency accuracy and resolution. As with many spectrometer systems, there is a large trade space in terms of different specific configurations for coherent dual comb spectrometers, some of which have been explored in past work [1-11]. We will discuss our specific experimental implementation, which has been targeted toward measuring gas spectra with a high spectral resolution and signal-to-noise through coherent averaging.

The basic concept of a coherent dual comb spectrometer is illustrated in Figure 1. In the frequency-domain picture, the two combs have slightly different tooth spacings so that their heterodyne signal leads to a comb in the rf domain. With appropriate care, the rf comb teeth can have a one-to-one correspondence with pairs of optical comb teeth. In the time-domain picture, the two comb sources produce pulse trains with different periods. As the LO pulses "walk through" the source pulses, the pulse-by-pulse overlap is digitized. The resulting interferogram is a down-sampled measurement of the source pulse after transmission through the gas; any gas absorption signature appears as a trailing free-induction decay.

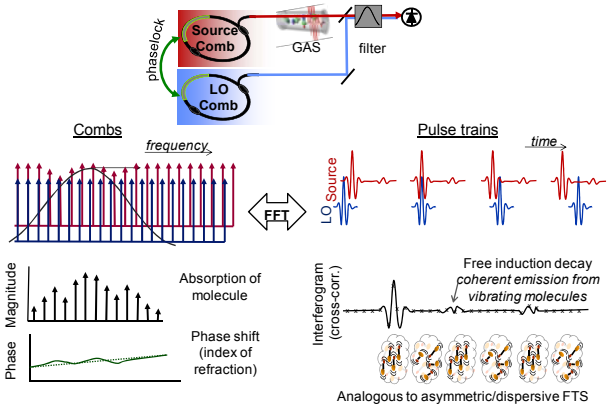

Figure 1: Simplified schematic of a dual comb interferometer. In the frequency comb picture, the heterodyne signal between the two offset combs is an rf comb. The amplitude and phase of the rf teeth reflect the amplitude and phase perturbations of the gas (if compared to a reference scan without gas.) In the time domain picture, the interferogram is measured as the LO pulses slowly walk through the signal pulses. The interferogram is measured repeatedly, at a rate equal to the difference in the comb repetition rates. The Fourier transform of multiple concatenated interferograms gives the rf comb shown on the left side. Note this picture is slightly oversimplified since it ignores the time and frequency domain response of the detector that will blur either the frequency or time domain pictures, but in a stable way.

There are many different configurations. The two comb sources can be combined before the sample for greater robustness against phase perturbations in the path but then the gas dispersion is not measured. The combs need not be phase-locked to demonstrate the basic operation, but then only a single, highly apodized interferogram is measured without the resolution of the comb and without any intrinsic accuracy. In our case and in Ref. [9], the two combs were tightly phase-locked to two cw lasers. Then the interferograms are stable over long time and one

measures an rf comb with discrete, well-defined rf teeth. One can also then average multiple interferograms for higher SNR. To achieve an accurate frequency axis, we also stabilize the two cw lasers to optical cavities and measure their absolute optical frequency against a self-referenced comb. In principle, then, the spectrum has the frequency accuracy of the cw lasers. In practice, the accuracy of any line measurement is limited by the signal-to-noise ratio and systematics. We are currently investigating the practical limits to the frequency accuracy of the system. If the instrument lineshape can be very well characterized, the system could probe molecular lineshapes under different environmental conditions.

2. System constraints

A dual comb spectrometer can measure a gas spectrum rapidly, over broad spectral bandwidth (compare to a tunable laser), with high resolution, high signal-to-noise ratio, and high accuracy. Of course, it cannot do all these things simultaneously. The most fundamental constraint is related to Nyquist sampling criterion, which limits the acquisition rate for an interferogram, f_{Int}, to $f_{Int} << f_{rep}^2/(\Delta\nu)$, where f_{rep} is the comb repetition rate and $\Delta\nu$ the spectral bandwidth. Shorter acquisition times are sometimes quoted, but these shorter times correspond to a single, highly apodized interferogram. The above relationship is an inequality; how well it must be satisfied is an interesting question and relates to the suppression of aliasing effects that might distort lineshapes.

This Nyquist constraint is not the only one. As is often the case with Fourier spectroscopy, the SNR is limited by the detector dynamic range at least for a bright comb source. Since the data is digitized at f_{rep} , some of the "tricks" used in conventional FTIR systems to achieve higher dynamic range are unavailable. For example, very high dynamic range detectors and sigma-delta analog-to-digital converters (ADC) are not available at these high bandwidths. Gain switching is also not possible at these speeds. However, it is possible to apply a differential chirp between the comb sources to stretch out the centerburst [9] to increase the dynamic range limit. The dynamic range limit translates to a maximum pulse power, since the pulse power determines the centerburst height. Therefore, there is a tradeoff between SNR and spectral bandwidth (because as the same pulse power is distributed over a larger spectral bandwidth the SNR per spectral channel decreases). This tradeoff can be alleviated by a divide and conquer approach where individual spectral slices are acquired separately, at the cost of system complexity [5, 7, 8].

Finally, there are other practical constraints, one of which is illustrated in Figure 2. As the comb spectrum is broadened, the power per spectral bin drops of course assuming the same amplifier, but more importantly, the variation in spectral amplitude grows, leading to an even greater the dynamic range challenge for detection.

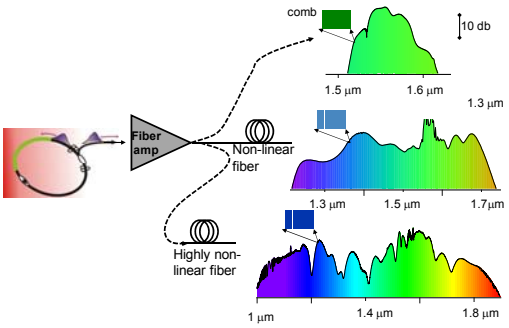

Figure 2: Spectra from a femtosecond fiber laser with varying degrees of spectral broadening. As the degree of spectral broadening is increased, the dual comb interferometer can access a wider spectral band, but the variations across the spectra become stronger and more challenging to deal with experimentally. For the octave-spanning spectrum shown at the bottom, the relative intensity noise can become a limiting factor to the SNR.

3. Example Data

The most extensive data set that we have acquired is for the rovibrational band of the overtone H-C stretch transition in Hydrogen Cyanide. This particular spectral band has a very nice overlap with the direct output of a femtosecond fiber laser, shown in the top panel of Figure 2. As discussed earlier, in our setup only the source comb is transmitted through the gas cell. Therefore, the signal has both an amplitude and phase component in the frequency domain. We measure this signal in small spectral bands to circumvent our detector dynamic range limit and then stitch together the complex spectra to generate the full rovibrational spectrum shown in Figure 3. An inverse Fourier transform of these data yields the full free-induction decay of this rovibrational band, as shown in the top right panel. Neither the complex frequency domain spectrum nor the free induction decay is particularly intuitive, but an intermediate time-frequency domain picture, illustrated in the bottom panel of Figure 3, is much more intuitive. It shows the initial pulse arrival, the bright white line at time zero, followed by the free induction decay of the molecules. For the high

time-resolution of this particular short time Fourier Transform, one can see the rotational recurrences in the P and R branches as the molecules rotate in and out of phase with each other.

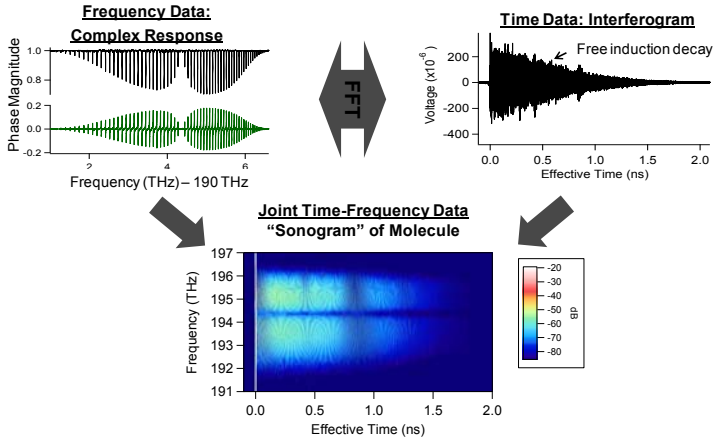

Figure 3: Example high SNR data from Ref. [7, 10] at a resolution of 200 MHz using an interleaved signal and reference pulse. (Top left) The complex frequency response, (Top right) the corresponding time-domain interferogram and (bottom) a short-time Fourier transform of the data.

While these data indicate the quality of the spectra possible with this system, the spectrum of HCN is perhaps not the most experimentally interesting and, furthermore, the spectral coverage was fairly limited in these experiments. We have therefore expanded the spectral coverage of the comb by modest spectral broadening in normal dispersion highly nonlinear optical fiber. The resulting spectrum is shown in the middle of Figure 2. This comb spectrum has higher amplitude structure but allows the spectrometer to probe a much wider range of molecules. We will present preliminary measurements of water at 1.4 μm and of methane at 1.6 μm. The latter is particularly interesting as a high resolution measurement of methane in this wavelength range can probe lineshape functions that are important for monitoring of climate change gases with high accuracy. Finally, the near infrared spectrum can be translated to the short-wave infrared through difference frequency generation. We will also present some preliminary measurements of methane spectra at 3.4 micrometers.

4. Conclusion

Coherent dual comb spectroscopy is an interesting new spectroscopic tool that is still being actively investigated in a number of laboratory setups. It can provide a broader spectral coverage than tunable laser spectrometers, and a higher accuracy, resolution , and SNR than many conventional FTIR systems. It has potential in at least two areas. If the technique can be successfully migrated further into the infrared, it could provide broadband sensitive detection of gases. If the technique can fully access the underlying accuracy and resolution of the frequency combs, it can provide high-resolution and accuracy gas spectra to complement those from state-of-the-art high resolution FTIRs .

5. References

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