Coherent Frequency Combs for Spectroscopy Spanning 3 to 5.2 µm

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Abstract: A tunable mid-infrared frequency comb was created via difference frequency generation. Pulses between 1 and 1.5 μ m were mixed to make idlers of 3-5.2 μ m. Two such combs were heterodyned at 5 μ m to show their coherence and potential for spectroscopy.

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Mid-infrared (MIR) laser sources are a powerful tool to study the spectroscopic fingerprints of various compounds important for atmospheric chemistry, trace gas detection, and fundamental science. The method of dual frequency comb spectroscopy [1,2] is a highly effective means to make rapid, high bandwidth, well-resolved accurate measurements, especially over a broad wavelength range in the mid-infrared. With this in mind, two MIR frequency combs spanning from 3 to 5.2 µm have been developed, each based on the difference frequency generation (DFG) of a near-infrared comb laser. While mid-infrared combs have been developed via a similar technique in this spectral region previously [3–6], this work aims to characterize the coherence and noise properties of these mid-infrared sources for their future implementation in precision dual-comb mid-infrared spectroscopy.



Fig. 1. Left: Schematic of the difference frequency generation apparatus. The diagram is explained in detail in the text. Right: The attainable spectral coverage using a 1 mm PPLN crystal. The gap at $4.2 \,\mu$ m is due to atmospheric carbon dioxide absorption in the path between the crystal and the Fourier transform spectrometer.

The near-infrared frequency combs used in generating the two mid-infrared combs were modifications of those referenced in [7] to allow for tunability from 3 μ m to 5.2 μ m. Two erbium fiber ring oscillators generated 1.5 μ m frequency combs at 100 MHz, which were the basis for the two DFG systems (see Fig. 1, left). The generated comb light from each was split into two branches, which produced the pump and signal used in DFG. Erbium-doped fiber amplifiers (EDFAs) were used to generate short pulses with high peak power at 1.5 μ m, which were launched into highly nonlinear fibers (HNLFs) to create low-noise dispersive waves at 1 μ m. These were the seeds for high-power ytterbium-doped fiber amplifiers (YDFAs), which produced 2 W at 1 μ m. The 1.5 μ m signal pulses were launched into short sections of HNLF; the pulses' chirp and peak power were catered towards generating spectral power between 1.3 and 1.5 μ m. The 1 and 1.3-1.5 μ m beams were combined in periodically-poled lithium niobate (PPLN) crystals (either 1 or 3 mm in length), with poling periods that allowed for the quasi phase matching of 1 and 1.3-1.5 μ m pump-signal pairs. Difference frequency generation produced several hundred microwatts of power over a wide band of wavelengths (see Fig. 1, right). Further gains can be achieved by increasing the pump power at 1 μ m, optimized

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beam focusing at the crystal, and careful tailoring of the broadened signal spectrum. (In a similar system, 0.5 W were achieved at 3 μ m [7].) At shorter wavelengths, the mid-infrared spectral width is limited by the spectral width of the signal near 1.5 μ m. At longer wavelengths, the mid-infrared spectral width is limited by the phase-matching bandwidth of PPLN. Coarse wavelength tuning is provided by the range of poling periods in the PPLN crystal. Finer tuning can be achieved by changing the temperature of the PPLN crystal.



Fig. 2. Left: Single (unaveraged) 5 μ m interferogram. Left inset: Spectrum of the unaveraged Fourier-transformed interferogram is shown in gray, and the transform of 16 averaged interferograms is shown in blue. Right: The relative intensity noise of the two MIR combs, as well as that of dark detector and the electronic measurement background.

Interferograms of the combined MIR combs were recorded using a liquid nitrogen-cooled mercury cadmium telluride (MCT) high-bandwidth (>100 MHz) detector. A sample interferogram from the overlapping of the two 5 µm combs is shown in the left frame of Fig. 2, along with the resulting transformed spectra. Here, the two combs' repetition rates were roughly 99.84 MHz, with a repetition rate difference of 290 Hz. The interferogram is a single shot, unaveraged, with both combs free-running and without any active stabilization. The inset shows the resulting spectrum from one unaveraged interferogram, and the noise reduction of 16 interferogram averages. The time-domain signalto-noise ratio of the interferogram is estimated to be approximately 25, within the 50 MHz Nyquist filtered bandwidth of the digital sampling system. The relative intensity noise (RIN) is shown at right in Fig. 2. Electronic noise is seen at low frequencies, but is native to the measurement setup and not the MIR source or the detector, as indicated by the electronic background. The large peak at 36 kHz is electronic noise from the power supply of one of the 1 µm amplifiers. The strong coherence and low RIN of this system show potential for immediate implementation in robust, high-sensitivity mid-infrared molecular spectroscopy.

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