Mid-Infrared Hyperspectral Microscopy with Broadband 1-GHz Dual-Comb Spectroscopy

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Abstract: We utilize a 1-GHz mid-infrared dual comb spectrometer for high speed and broadband hyperspectral imaging. The system covers 1000 cm^{-1} at $\lambda_c = 3.4$ μm with 13.47 kHz acquisition rate and 4 μm spatial resolution. © 2023 The Author(s)

Mid-infrared spectrometers covering the $\sim 3-12\mu$m wavelength range give direct access to many fundamental molecular and lattice vibrations that constitute the low energy landscape of solids and biological tissue. When combined with an imaging system, mid-infrared microscopy has enabled the development of new technologies for label free chemical imaging [1].

A goal of hyperspectral microscopy is to realize the combination of high speed spectrometers operating at broad frequency bandwidths, together with microscopes operating at sub-micron spatial resolution. Imaging below the diffraction limit has been demonstrated using near-field microscopy [2], or by leveraging the photothermal and optical nonlinear response of sample materials [3–5]. The dual frequency comb has emerged as a platform for broadband and high speed imaging [6, 7]. However, since its introduction a mid-infrared spectrometer occupying the target intersection of speed (exceeding $\sim$ 10 kHz) and bandwidth (tens of terahertz) still needs to be realized in imaging applications. Recently we demonstrated a broadband 1-GHz mid-infrared dual-comb spectrometer [8, 9]. In this work, we exemplify the utility of this light source for broadband and high speed spectroscopy in a mid-infrared imaging microscope. The system covers a bandwidth of $\sim$ 30 THz (1070cm^{-1}) at $\lambda_c = 3.4$ μm with $\sim$ 74 μs single shot acquisition time, or equivalently a 13.47 kHz interferogram acquisition rate.

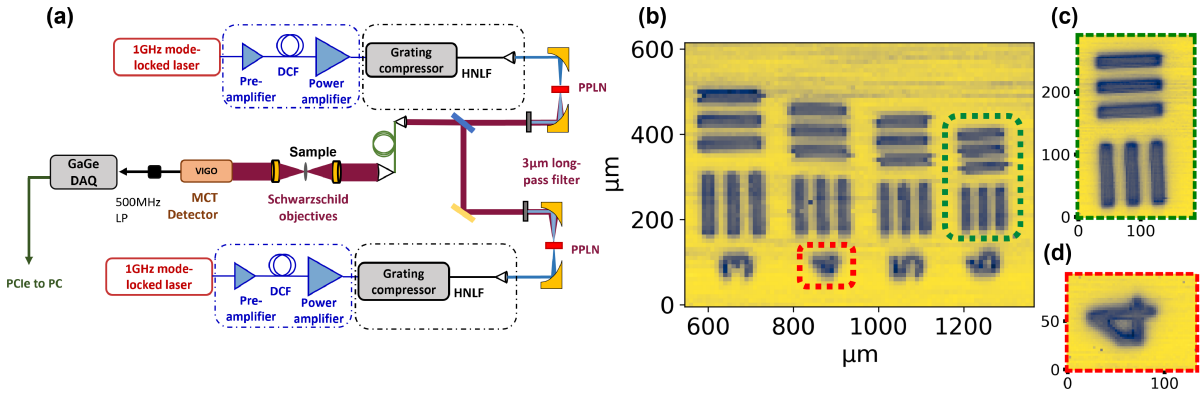

Fig. 1. (a) Diagram of experimental setup [8] (b) Coarse 10 μm / pixel image of the USAF target generated by integrating the absorbance across the wavelength window in Fig. 2. b. (c) 2.5 μm / pixel image of element six (d) 2.5 μm / pixel image of the number "4".

The broadband mid-infrared frequency combs are generated from a simple design shown in Fig. 1. a. using single-branch intrapulse difference frequency generation in periodically poled lithium niobate (PPLN). The pump is generated by spectrally broadening the amplified output of a 1-GHz mode-locked oscillator in anomalous dispersion highly nonlinear fiber (AD-HNLF). The length of HNLF is chosen to meet the pulse at its soliton fission point to obtain a few cycle and several nanojoule ($\sim$ 10 fs, 3.3 nJ) pulse. In the single-branch design, the conversion efficiency in a 1 mm thick fan-out PPLN reaches between $0.1-0.2$ % resulting in $3-6$ mW of average

power, and whose peak wavelength can be continuously tuned to cover the $3-5\ \mu$m window. The mid-infrared light is collinearly focused onto the sample with an estimated $\sim 4\ \mu$m spot size, and re-collimated with 0.58 NA reflective objectives. Spectral images are gathered by raster-scanning the sample through the beam focus.

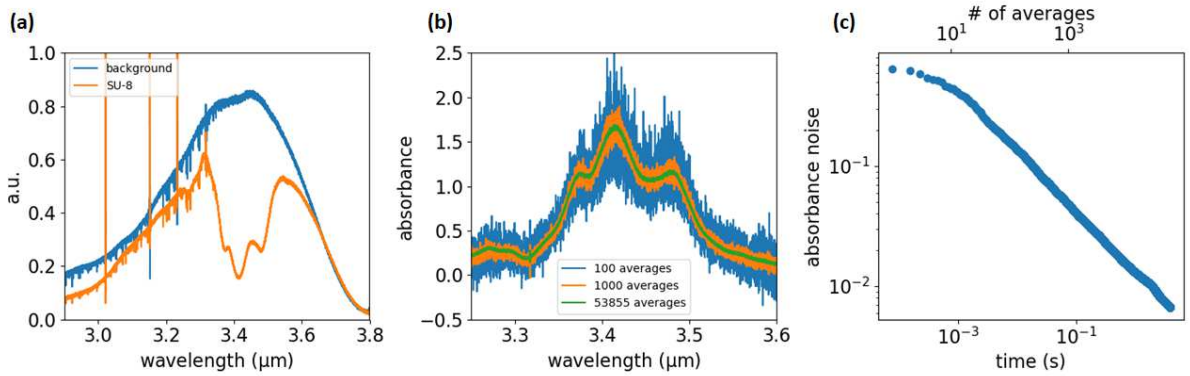

Fig. 2. (a) Raw spectra for the background and on an SU-8 bar (b) Absorbance of a bar of SU-8 photoresist taken at 100 (7.4 ms), 1000 (74 ms) and 53,855 (4s) averages. Resolution is 1 GHz or 0.03 cm^{-1}. (c) The absorbance noise as a function of averaging time.

As an initial characterization, we spectrally image a USAF test pattern of SU-8 photoresist on a 0.5 mm silicon wafer. The images in Fig. 1. are generated by integrating the absorbance after 1000 averages (the orange curve in Fig. 2. b.), which results in a 74 ms averaging time per pixel. A coarse image with 10 μm pixel size is shown in Fig. 1. b., with higher resolution images at 2.5 μm pixel size shown in Fig. 1. c-d. for element six and the number "4". Fig. 2. a. shows the raw spectrum for the background (taken between the bars) and on an SU-8 bar. Cascaded $\chi^{(2)}$ in the PPLN crystal causes the offset frequency of the near-infrared pump to appear in the mid-infrared, resulting in beat notes that overlay the spectrum. However, the fundamental mid-infrared combs are offset free. The broad absorption at $\lambda_c = 3.4\ \mu$m is shown in Fig. 2. b, with Fig. 2. c. showing the calculated absorbance noise as a function of averaging time.

In conclusion, we demonstrate a high speed dual-comb mid-infrared microscope with >10 kHz acquisition rate and covering $\sim 1000\ \text{cm}^{-1}$ of optical bandwidth. In this work the interferograms are not truncated, thereby giving the full 1 GHz resolution of the frequency combs. However, for broad absorption features the signal to noise can likely be improved with apodization, allowing the system to operate closer to single shot. Applied to this experiment, the averaging number can likely be under 100 (7.4 ms). Lastly, for such broad features, it is interesting to consider even higher repetition rates. For instance, a 10 GHz system that spans the same Nyquist window would achieve an acquisition rate of 1.37 MHz, pushing the single pixel averaging time below the 100 μs regime.

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