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Free-form dual-comb spectroscopy for compressive sensing and imaging

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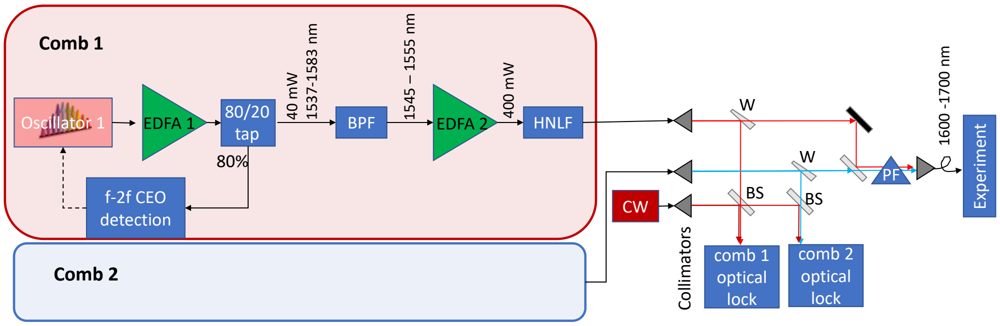

Figure S1: Optical setup. Each comb is shown as a shaded box. The components for comb 2 duplicate comb 1 but are not shown. The free-space section that combines the CW laser and comb light avoids larger slow phase and timing drifts that can occur in a fully fiber-based system. The two comb outputs at the far-right side are filtered by a free-space prism filter (PF) and then coupled back into fiber for delivery to either the gas cell setup or the camera setup. Oscillator: linear Er+ fiber cavity with a semiconductor saturable absorber mirror, pumped by one 1480 nm laser diode. EDFA 1: Erbium-doped fiber amplifier pumped by two 980 nm laser diodes. f-2f CEO detection: Carrier-envelope offset frequency accomplished by broadening in a highly nonlinear fiber followed by doubling of the 1960 nm supercontinuum light in periodically poled potassium titanyl phosphate (PPKTP), and photodetection. BPF: 10 nm optical bandpass filter. EDFA 2: Erbium-doped fiber amplifier pumped by three 980 nm laser diodes. HNLF: highly nonlinear fiber for spectral shaping. CW: narrow band continuous wave laser. W: CaF_2 wedge (7% reflection). BS: 50:50 beam splitter. Fiber paths are shown as black lines, fiber-to-free space collimators as grey triangles, and free-space paths as colored lines.

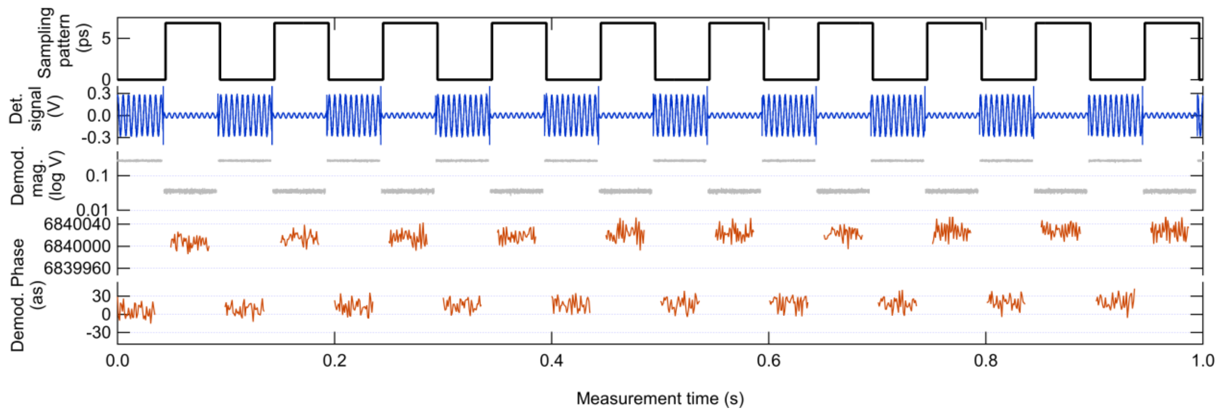

Figure S2: Accuracy of relative pulse delay stepping illustrated in recurrence sampling mode where the relative pulse delay ΔT_{RPD} is jumped between 0 ps and 6.84 ps to alternatively probe the centerburst and a methane recurrence signal. The commanded ΔT_{RPD} (sampling pattern, black trace) leads to the modulated heterodyne signal (blue trace), which is IQ demodulated to retrieve the signal magnitude (grey trace) and phase (red trace). The demodulated phase is unwrapped, converted to time units using a 2π optical carrier period of $1/(184 \text{ THz}) = 5.5 \text{ fs}$, and added to the commanded ΔT_{RPD} to obtain the demodulated phase timing (red trace). It has about 10 as 1-σ precision and 40 as peak-to-peak wander, which puts an upper limit on most (e.g. uncorrelated or non-cycle slip) errors in the actual ΔT_{RPD}. (Cycle slips would appear as a slow change in the signal magnitude as the relative pulse delay moves off the centerburst or recurrence and are not observed). Over much longer time periods of minutes, the phase can have a peak-to-peak wander of 150 attoseconds due to drifts in optical path lengths associated with the phase locking of the combs.

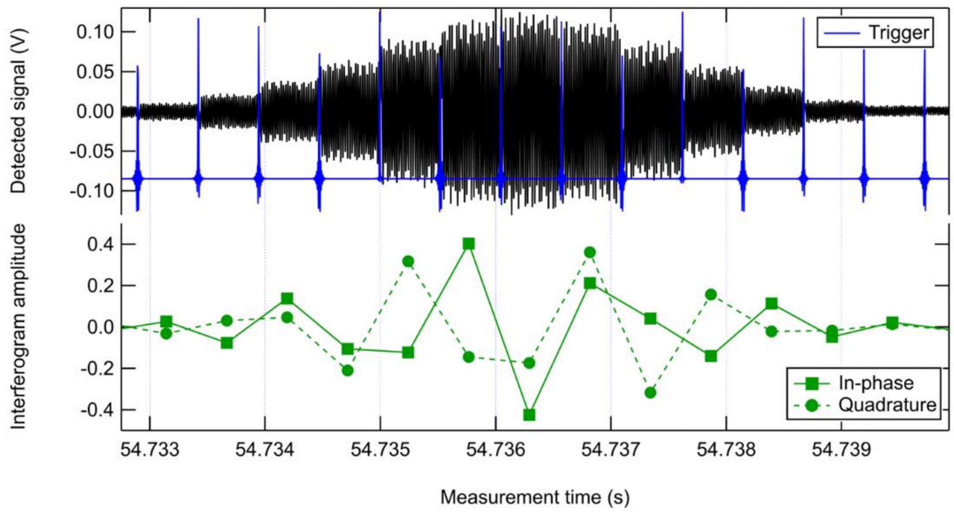

Figure S3: Example of measured time-domain signal focusing on the centerburst portion of the interferogram. At each $t_{dwell} + t_{dead}$ = 0.5 ms long interval, the relative pulse delay is fixed, and the signal is a sinusoid at f_{if} = 50 kHz (top graph). A trigger signal generated by the digital comb control hardware (shown in blue) indicates the initiation of a step and the start of a dead time t_{dead} used to mask out transients caused by actuator responses. The signal is demodulated in phase and quadrature, and then averaged over t_{dwell} to give one in-phase and one quadrature value of the interferogram (bottom plot). If the step size is equidistant across the entire interferogram, the resulting demodulated signal is identical to one acquired with traditional DCS. For example, a full step scan over 208332 steps at $\Delta T_{RPD} =$ 30 fs and t_{dwell} = 0.5 ms requires about 109 s and achieves a spectral SNR of 630, which is the same SNR reached using coherent averaging over the same 109 s acquisition time in a traditional DCS measurement at an offset repetition frequency of Δf_r = 130 Hz (in traditional DCS, the choice of Δf_r doesn't impact the SNR; it just needs to be sufficiently small such that $f_r^2 / \Delta f_r > \Delta \nu$). The t_{dead} used here is much shorter than the 0.5 ms used in the main manuscript's compressive sensing measurements, which was chosen to accommodate much larger steps in ΔT_{RPD} .

a)

b)

c)

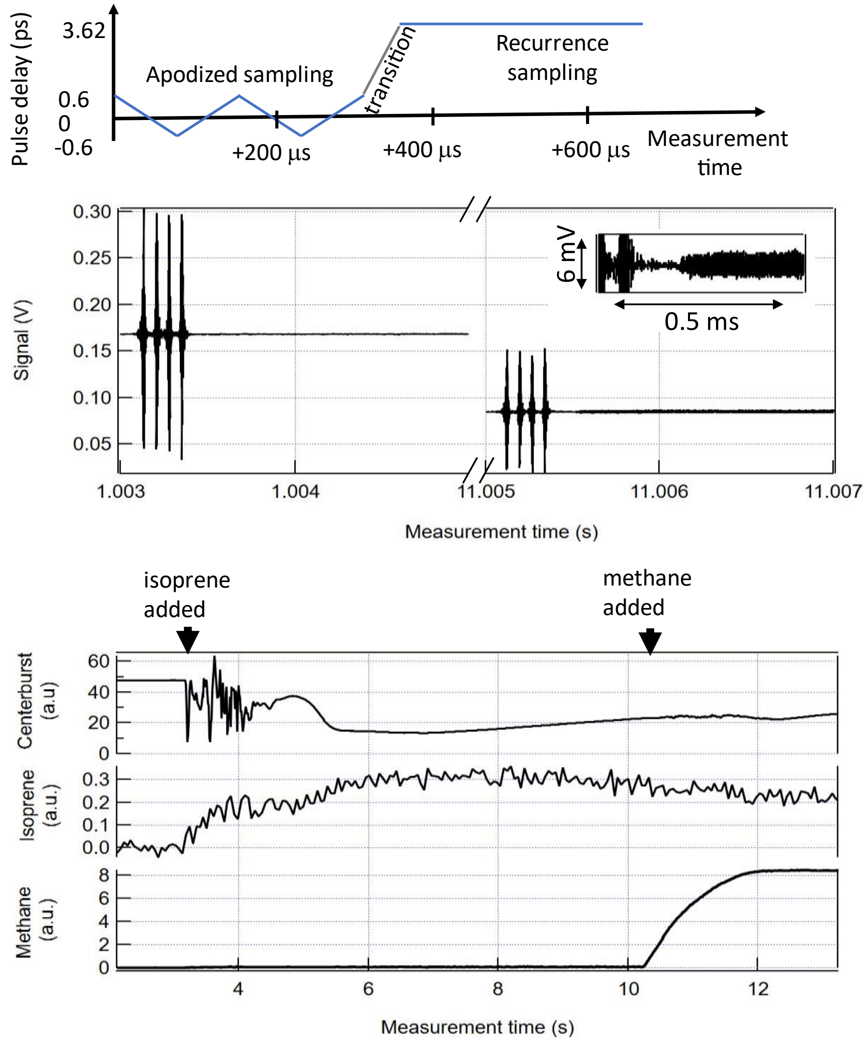

Figure S4: Example of combined real time apodized and recurrence sampling probing a mixture of isoprene and methane. a) Sampling pattern consisting of four consecutives real time apodized scans, in this case using offset repetition frequencies for simplicity rather than a stepped approach, followed by a jump to a relative pulse delay T_{RPD} = 3.62 ps corresponding to the first recurrence for methane. The measurement pattern has an update rate of 10.6 ms, one apodized scan over the centerburst is 72 μs (4 such scans are taken), the recurrence, after the jump to T_{RPD} = 3.62 ps, is probed at f_{IF} = 300 kHz. b) The measured time domain signal. At the beginning of the experiment neither isoprene nor methane is present in the gas cells (left half). Eleven seconds into the measurement both gases are introduced (right half). Inset: the expanded signal with both gases present showing the slightly distorted centerburst, due to the isoprene, and the recurrence signal from the methane gas, the oscillations to the right. c) The three main extracted signals are the centerburst height, which is used for normalization of the recurrence signal, the retrieved isoprene concentration based on a spectral analysis from the four consecutive apodized scans (by Fourier transformation of the appropriate time-sliced data) and the retrieved methane concentration based on the demodulated magnitude of the recurrence signal. Overall signal variations, here mostly due to some condensations of cell windows when liquid isoprene is filled in and starts evaporating, can easily be calibrated out from the recurrence signal by taking the centerburst into consideration. The spectral analysis to obtain the isoprene concentration is less affected and a baseline correction can be applied.

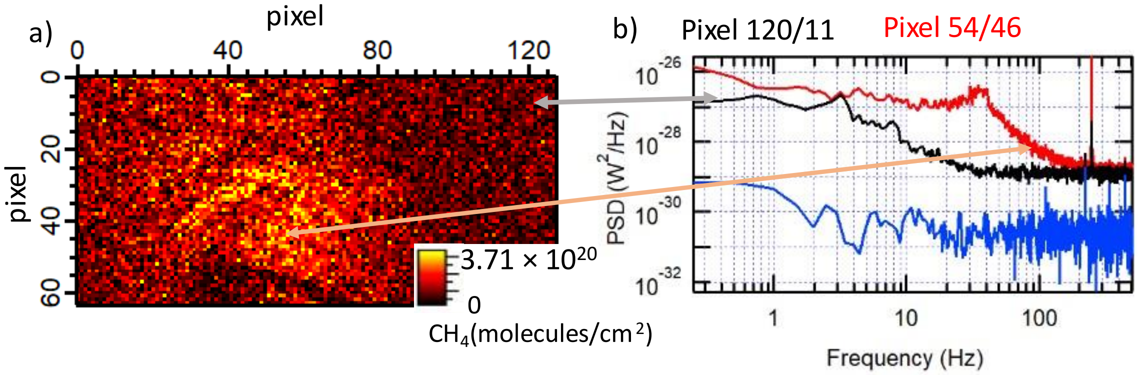

Figure S5: Time domain analysis of the recurrence species-selective imaging of Figure 5. a) Example frame showing the demodulated magnitude scaled to methane concentration. b) Power spectral density (PSD) of the raw signal scaled to received power for a pixel within the methane plume (red, top trace), outside the plume (black, center trace), and with the dual comb light blocked (blue, bottom trace). The increase in the noise floor from ~ 2 x 10^{-31} W^2/Hz to ~ 10^{-29} W^2/Hz in the PSD amplitude when the FPA is illuminated is attributed to relative intensity noise (RIN) on the frequency combs. For the pixel within the plume, turbulence effects within the methane flow lift the PSD even above this RIN limit, exhibiting a knee at ~ 30 Hz with a slow decay to 200 Hz, depicting that f_{IF} should be set to higher frequencies for better SNR. The spike at 250 Hz is the frequency difference f_{IF} between the combs' CEO lock frequencies; the magnitude of this 250 Hz signal is shown in panel a). It is interesting to contrast the recurrence species-selective imaging shown here with a cw differential absorption-based imager. The biggest advantage of the cw laser is sensitivity. For an imaging system SNR will typically be limited by pixel bit depth and a cw laser is able to more efficiently use this range. For the 12-bit camera and nearly saturated methane sample shown here we estimate that the cw laser would have a 17× higher sensitivity. However, the speed is most often limited by the camera frame rate making it hard to leverage this sensitivity for increased measurement speed but there is an SNR benefit. For a single gas species where a cw laser is readily available it remains the most sensitive approach. However, if one wants a single system that could be tuned to many different species the comb-based recurrence sampling is still an attractive option for high speed imaging.