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Supplementary Materials for

Infrared electric field sampled frequency comb spectroscopy

Abijith S. Kowligy*, Henry Timmers, Alexander J. Lind, Ugaitz Elu, Flavio C. Cruz,
Peter G. Schunemann, Jens Biegert, Scott A. Diddams*

*Corresponding author. Email: abijith.kowligy@gmail.com (A.S.K.); scott.diddams@nist.gov (S.A.D.)

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Reference (*61*)

Section S1. Noise in EOS detection

Strong optical filtering of the SFG output of the EOS crystal has conventionally led to shot-noise limited performance. In this case the total filtered optical power in the 1300-nm band is P = 200 µW, half of which is incident on each port of the BPD, which has a responsivity of 0.95 A/W. The theoretical photo-current due to shot-noise is $I_{sh} = \sqrt{2qI\Delta f} = 5.7$ pA/Hz$^{1/2}$. As this fundamental noise cannot be subtracted in balanced detection, it is added in quadrature, resulting in an 8 pA/Hz$^{1/2}$ input to the trans-impedance amplifier (TIA) , with gain of 25×10^3 .The predicted rms noise voltage after the TIA in 50 MHz bandwidth is then 1.4 mV. However, we observe 5 mV noise voltage on the oscilloscope (measured without an infrared input to the EOS crystal), corresponding to 5.5 dB excess noise. We speculate that the inadequate common-mode rejection for 50 MHz electrical bandwidth in the BPD and extraneous electronic noise in the TIA are the leading candidates for this excess noise. Nonetheless, single-shot time-domain SNR of 40 in 50 MHz detection bandwidth is observed for the electric field measurement. The frequency domain SNR is 1 Hz$^{1/2}$ per comb tooth, corresponding to a sensitivity per comb tooth of 100 pW/Hz$^{1/2}$, with 50 µW MIR power reaching the EOS crystal with 50 THz of optical bandwidth. Equivalently, we can write this sensitivity as 100 photons per comb tooth per pulse. Moreover, the figure-of-merit (FOM, $M \times$ SNR), accounting for the number of comb-teeth (M) is comparable to other published dual-comb spectroscopy results [1]. More specifically, for the OP-GaP IR comb, corresponding approximately to 7.5×10^5 comb teeth, the FOM is 6.75×10^5, comparable to the the state-of-the-art MIR DCS performance [2] around 5 µm that also utilized active phase-correction. The time-domain single-shot SNR of 40 is distributed across M=7.5×10^5 comb teeth, thereby decreasing the spectral domain SNR, requiring minute-scale averaging for spectroscopic data. The averaging times can reduced by increasing the power from the nonlinear crystal and operating at the shot-noise limit.

Section S2. Noise equivalent absorption in dual-comb EOS

In the broadband spectra obtained from OP-GaP, the frequency domain signal-to-noise ratio is > 76 in 110 minutes of averaging, corresponding to peak minimum absorption for the 15-cm-long gas cell to be, $\alpha_{min} = 8.6 \times 10^{-4}$ cm^{-1}. We note that the acceptance bandwidth of the EOS detection, determined mathematically by the convolution of the NIR sampling pulse duration and the phase-matching bandwidth, is in excess of 60 THz in our system. With the figure-of-merit described in the previous system, our empirical results agree well with the predictions described by Coddington et al. [1].

Section S3. Atmospheric water vapor in the 585- to 630-cm^{-1} band

Due to the expansive phase-matching bandwidth and high sensitivity provided by EO detection, we measured the absorption fingerprints of atmospheric water vapor in the 585–630 cm^{-1} (15.9–17 µm) that is inaccessible via state-of-the-art high-speed ($>$ 50 MHz) infrared photo-detectors. Moreover, monitoring the absorption of CO_2 and H_2O in this band is important for environmental monitoring [3]. Quantitative agreement is observed between the line centers and line intensities to modeled atmosphere in this spectral region.

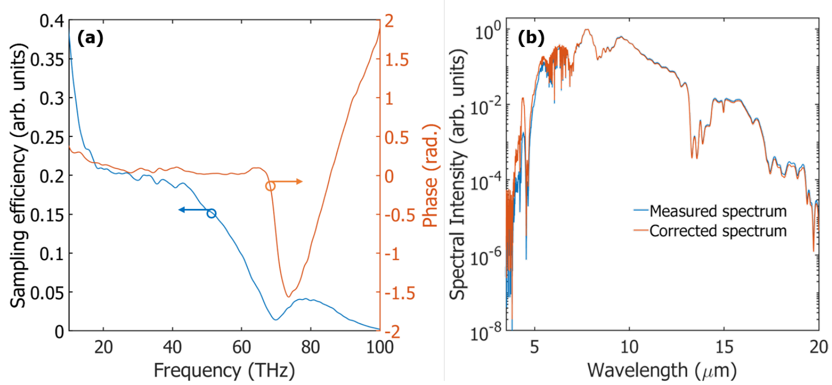

Fig. S1. EOS response function. (a) The amplitude and phase of the response function for electro-optic sampling with the 10-fs near-infrared pulse and the 30-µm thick GaSe crystal is shown, calculated using the formalism developed in [4]. Due to the finite spectral bandwidth and pulse-duration in the sampling pulse, the higher frequencies are sampled less efficiently. (b) The measured spectrum and the response-function-corrected spectrum generated in orientation patterned gallium phosphide is shown with a 4 cm^{-1} resolution. As expected the higher frequency components near 4 µm are corrected in amplitude owing to the lower sampling efficiency.

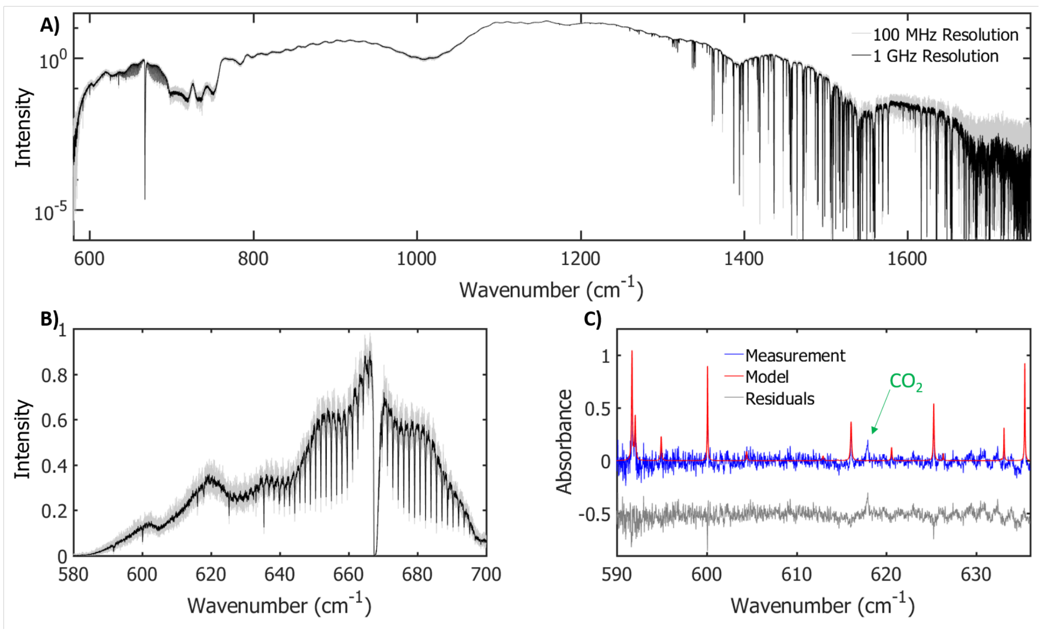

Fig. S2. Atmospheric water vapor absorption in the 585- to 630-cm^{-1} band. (a) The instantaneous bandwidth of the measured signal extends from 580–1600 cm^{-1}, with the detection occurring in a 150-µm thick GaSe crystal. The averaging time is 110 minutes. The 100 MHz native resolution is shown in gray, with the numerically apodized 1-GHz resolution data shown in black. The SNR increases by a factor of $\sqrt{10}$ in the 1-GHz resolution data. (b) The transmission of the OP-GaP frequency comb through atmospheric carbon dioxide and water vapor in the 14.3–17.1 µm spectral region, shown with high-SNR. (c) Quantitative comparison with a modeled atmosphere shows good agreement. The residuals are offset from zero.

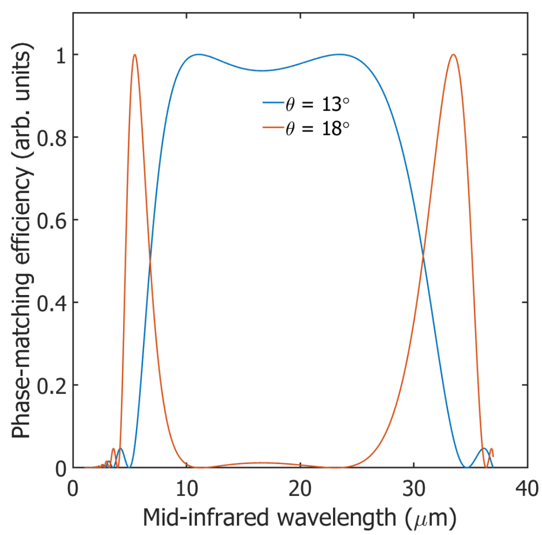

Fig. S3. Phase - matching curves for SFG in GaSe.
The phase-matching curves for Type-II sum-frequency generation in 30-µm-thick gallium selenide are shown as a function of internal angle. In the experiment, the angles are adjusted to 18 degrees or 13 degrees to increase the peak-to-peak electro-optic signal depending on whether the mid-infrared light is generated in periodically poled lithium niobate (PPLN) or orientation-patterned gallium phosphide (OP-GaP), respectively.