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**Title:** Ultrafast-laser-absorption spectroscopy in the mid-infrared for single-shot, calibration-free temperature and species measurements in low- and high-pressure combustion gases

**Authors:** Ryan Tancin, Christopher Goldenstein

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# Ultrafast-laser-absorption spectroscopy in the mid-infrared for single-shot, calibration-free temperature and species measurements in low- and high-pressure combustion gases

This supplementary document outlines several key details and considerations for acquiring high-fidelity, ultrafast-laser-absorption spectroscopy measurements. Included is a discussion on the effect of different spectral interpolation methods, as well as practical considerations for imaging ultrashort pulses in the mid-infrared.

## 1. BINNING INTERPOLATION

The high dispersion per pixel of ULAS measurements requires that absorbance spectra be simulated at a higher resolution than the measurement. Interpolating a spectrum between a high resolution simulation and the lower resolution measurement must be done in a manner which mirrors the physical discretization of the spectra by the camera's focal plane array (FPA).

Figure S1(a) illustrates the discretization of a spectrum of $I_t$ by the camera's FPA. A laser pulse which has been dispersed and instrument broadened by a spectrograph approaches the camera's FPA with a continuous intensity spectrum. When the pulse hits the FPA, the light is collected by individual pixels, in effect, spatially binning the signal according to the pixel's physical dimensions. At the end of the camera's integration time, the camera reads out a single value for each pixel, which corresponds to the spatiotemporally averaged incident intensity. In the spectral-fitting routine, $I_{t,HR,conv}$ is the analog of the real-world continuous intensity spectrum. This spectrum must be down-sampled to the experimental frequency axis using the same binning method as the FPA, by calculating the average intensity across a given pixel.

The difference between "binning interpolation" and linear interpolation is illustrated in Fig. S1(b-c). The error between interpolation methods is generally proportional to the second derivative of the spectrum being discretized, which is shown for reference in Fig. S1(d). In essence, the differences between method of interpolation become significant when the intensity varia-

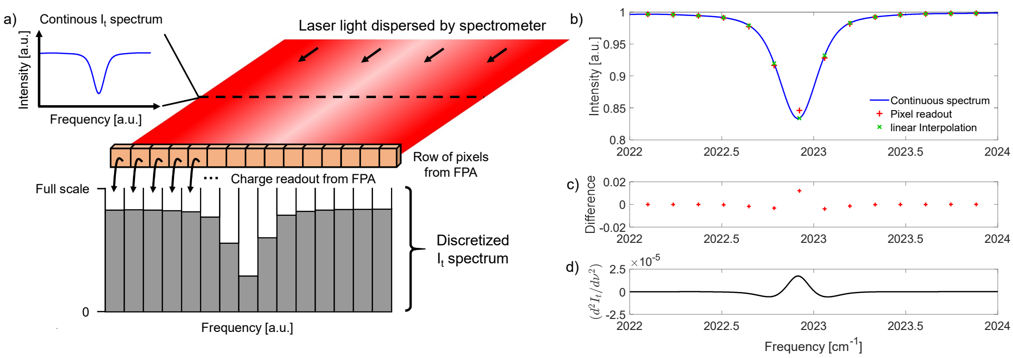


**Fig. S1.** A schematic illustrating the discretization process of an FPA, and the fidelity of binning and linear interpolation methods on mirroring this process. a) The discretization of a transmitted intensity spectrum by a row of pixels on the FPA. b) A plot comparing a continuous spectrum of $I_t$ with both a binning and linear down-sampling interpolation applied. c) Error (difference) between the binning and linear interpolation methods. d) Second derivative of transmitted intensity spectrum.

tion across a pixel cannot be assumed to be linear, because for linear variation, both a linear interpolation and the spatial mean value will be equal.

## 2. IMAGING ULTRAFAST LASER PULSES IN THE MID-IR

During the course of the development and initial applications of the ULAS diagnostic, several significant practical considerations for imaging ultrafast, mid-IR laser pulses were identified and are discussed below.

### A. Camera saturation

The IR camera used for ULAS experiments (Telops FAST-IR 2K) [1–4] has two modes of saturation which are relevant to ULAS measurements. The first is saturation of the photocurrent generation process in the FPA. If the signal flux (i.e., signal counts per unit time) is $\geq \approx 5000$ counts/$\mu$s (this value may range from 3000 to 6000 counts/$\mu$s for a given camera), additional incident photons will not be converted to signal counts. This is problematic in situations with short exposure times and high signal. For ULAS measurements, this was mitigated by using an integration time $\geq 4$ $\mu$s, though, 5 $\mu$s was used to provide a margin of safety.

The second relevant saturation mode corresponds to premature saturation of the number of possible signal counts. The typical response of the camera is between 0 and 65535 counts (i.e., 16-bit resolution), however, when imaging ultrafast pulses, saturation is observed at $\approx 18\%$ of its full range ($\approx$ 12,000 counts). It is currently hypothesized that this saturation mode is caused by depletion of charge carriers in the FPA and is distinct from the photocurrent saturation mode. This saturation mode is avoided by limiting signal counts to $\leq$ 10,000 counts, thus, allotting 2,000 counts of buffer for shot-to-shot laser fluctuations, drift in the time-averaged laser power, and emission during flame measurements.

Both of these saturation modes, even when avoided, have negative effects on ULAS measurements. Most notably, premature saturation limits the SNR of acquired spectra. Given that pixel noise is constant in magnitude, limiting signal counts to 12,000 reduces the potential SNR, and therefore detection limit, by a factor of five. Photocurrent saturation also limits SNR by limiting the camera's integration time and therefore the ability to reduce the contribution of the thermal background. For example, at an integration time of 5 $\mu$s the ambient thermal background typically contributes $\approx$ 1500 signal counts to acquired spectra, meaning that only 8,500 counts are left for recording laser intensity.

### B. Shot-to-shot repeatability

Shot-to-shot repeatability, in both the spectral shape of the laser intensity, as well as the camera's response is vital for acquiring high-fidelity, single-shot ULAS measurements. While there are complex spectral variations in the baseline laser intensity (see Fig. S2(a)), the shape of the spectrum is essentially constant with time, and mainly varies by a scalar multiple which enables

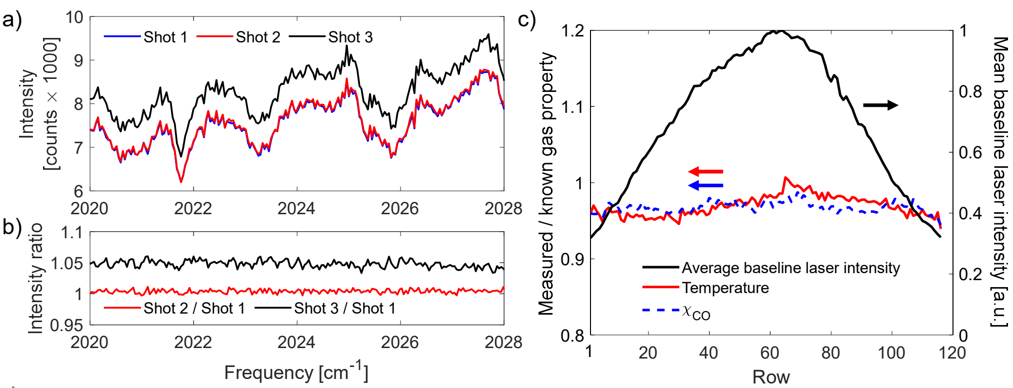


**Fig. S2.** (a) Three shots of $I_0$. Shot 1 and Shot 2 were recorded consecutively, and Shot 3 was recorded $\approx$ 6 minutes later. (b) Shots 2 and 3 are normalized to Shot 1 to highlight the stability in spectral shape, and suitability of a linear baseline correction. (c) The average value for $I_0$ is plotted as a function of FPA row, along with gas properties inferred from single-shot, single-row spectra to demonstrate the linear response of the IR camera to ultrafast laser pulses.

ULAS to provide high-fidelity single-shot measurements. A small linear term was added to the scalar baseline correction which yielded further improvement. The stability of the baseline laser intensity between subsequent laser shots and over the course of $\approx$ 6 minutes is illustrated by Fig. S2(a-b).

### C. Pixel linearity

The linearity of the InSb FPA was investigated to support the accuracy of ULAS measurements and assess the potential for 1D intra-pulse measurements of gas properties. Linearity was tested by processing and extracting gas properties from an image of transmitted intensity data which had pronounced intensity variation across its rows (the spatial dimension). A dataset from the gas cell experiments described in Section 4.2 was selected since the gas conditions were spatially uniform. Data was acquired at 10 bar using the grating with 300 lines/mm and processed according to the methods described in Section 3. The mean baseline laser intensity is shown in Fig. S2(c), which illustrates that the mean intensity varied by nearly a factor of three across rows of pixels. Gas properties inferred from single-shot, single-row spectra are also plotted as a function of row. The temperature and CO mole fraction varied by only 6%, indicating that the camera's FPA responded linearly to the intensity of the ultrafast laser pulses. Further, this demonstrates the ability of ULAS to acquire 1D-resolved, single-shot measurements of gas properties, and supports the accuracy of the 1D T and CO measurements acquired in an HMX flame (shown in Section 5.3 of the manuscript).

## REFERENCES


1. R. J. Tancin, Z. Chang, V. Radhakrishna, M. Gu, R. P. Lucht, and C. S. Goldenstein, "Ultrafast laser absorption spectroscopy in the mid-infrared for measuring temperature and species in combustion gases," in *AIAA Scitech 2020 Forum*, (2020), p. 0517.
2. R. J. Tancin, Z. Chang, M. Gu, V. Radhakrishna, R. P. Lucht, and C. S. Goldenstein, "Ultrafast laser-absorption spectroscopy for single-shot, mid-infrared measurements of temperature, CO, and $CH_4$ in flames," Opt. Lett. **45**, 583–586 (2020).
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