

## Tomographic laser absorption imaging of combustion species and temperature in the mid-wave infrared

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**Abstract:** In this work, laser absorption spectroscopy techniques are expanded in spatial resolution capability by utilizing a high-speed infrared camera to image flow fields backlit with tunable mid-wave infrared laser radiation. The laser absorption imaging (LAI) method yields spectrally-resolved and spatially-rich datasets from which quantitative species and temperature profiles can be generated using tomographic reconstruction. Access to the mid-wave infrared (3–5  $\mu$ m) enables imaging of fuels, intermediates, and products of combustion in canonical small-diameter flames (< 1 cm). Example 1D measurements and 2D reconstructions of ethane (3.34  $\mu$ m), carbon monoxide (4.97  $\mu$ m), and carbon dioxide (4.19  $\mu$ m) in an axisymmetric laminar flame are presented and discussed. LAI is shown to significantly enhance spatio-temporal data bandwidth (~400 simultaneously sampled lines-of-sight) and resolution (~50  $\mu$ m) compared to other tomographic absorption spectroscopy techniques, and with a simplified optical arrangement.

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#### **References and links**

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#### 1. Introduction

Tomographic absorption spectroscopy (TAS) has become an important optical approach to study high-temperature gas dynamics and reactive flows, including turbulent and laminar flames [1]. Laser absorption provides for a relatively straightforward, quantitative relationship to gas properties. In addition, calibration-free absorption techniques that exploit rapid laser tunability enable application to harsh combustion flows that often convolute other imaging techniques [2]. Advancements in TAS over the past decade, including several novel optical arrangements, have yielded significant improvement in spatial and temporal resolution. These advancements have mostly been confined to the near-infrared wavelength domain, where a multiplicity in laser beams and detectors comes at moderate cost and complexity, but the accessible combustion species are few (e.g.  $H_2O$ ) at the pathlength scales (~cm) of most flames [3–8]. By contrast, the mid-wave infrared provides for sensitive absorption spectroscopy of numerous gas species including fuels, intermediates, and products of combustion at their fundamental vibrational frequencies/wavelengths. However, optical methods for tomographic absorption spectroscopy in the mid-infrared have remained relatively rudimentary (e.g. mechanically translating lineof-sight) [9–13], limiting spatial and temporal data collection rates and utility in combustion studies. In this paper, we describe a novel tomographic laser absorption imaging method using mid-wave infrared optics and a high-speed camera that significantly enhances spatio-temporal data bandwidth (i.e. mapping a 2D flow-field in seconds vs. minutes or hours) and enables quantitative analysis of several combustion species and temperature in small-diameter flames utilized for fundamental combustion investigations.

#### 2. Method

#### 2.1 Optical setup

The optical arrangement for tomographic laser absorption imaging (LAI) involves one or more high-speed infrared cameras that image a flow-field backlit with tunable mid-wave infrared laser radiation. The simplest configuration, suitable for tomography of axisymmetric flows, includes a single laser and camera, shown in Fig. 1. The laser beam is expanded with a concave lens, similar

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Fig. 1. (*Left*) Arrangement of lasers, mirrors, Bunsen-style flame, and IR camera. (*Right*) Normalized beam profiles measured by IR camera for 200-by-2 pixel regions of interest.

to the fan beam method [5–7], and recollimated with a convex lens to pass through the flow of interest (at a size large enough to capture the entire width or half-width of the absorbing medium) and spectrally filtered to isolate the laser radiation. The enlarged beam ( $d \approx 20$  mm) is then focused onto a 2D focal plane array comprising many detector pixels that effectively represent unique optical lines of sight from the collimated source sampled simultaneously. In this work, we use a high-speed IR camera (Telops TS-IR-MW) with a 640-by-512 pixel CCD array and mercury-cadmium-telluride (MCT) photodetector. The maximum frame rate of the camera is 107 kHz, which requires a significant reduction in the number of active pixels and integration time. For the experiments reported here, the frame size is set to 200-by-2 pixels, capturing a transverse slice of an axisymmetric flow, and the frame rate is 40 kHz with an exposure time of 3  $\mu$ s. The corresponding beam intensity profiles for the different lasers used in this study are shown in the right of Fig. 1. Each beam exhibited some diffraction patterns as imaged, but a relatively stable near-Gaussian section could be attained.

For demonstration of the LAI method, three tunable narrow linewidth mid-infrared lasers were used to measure absorption profiles of  $C_2H_6$ , CO, and CO<sub>2</sub>, respectively, in the radial and vertical directions to characterize the thermochemical structure of a laboratory Bunsen-style  $C_2H_6$ -air flame, pictured in Fig. 2. For each experiment, only one laser and bandpass filter combination is used at a time. The burner is mounted on a vertical translation stage controlled by a stepper motor with a linear translation of 5 mm/sec (5  $\mu$ m per step) to provide vertical resolution. The partially premixed flame was controlled via thermal-based mass flow controllers (MKS MFC GE50A) with flow rates of 179 sccm  $C_2H_6$  and 1391 sccm air, corresponding to a fuel-rich mixture with a fuel-air equivalence ratio of  $\phi = 2.14$  and exit velocity of 2.66 m/s. The exit of the stainless steel burner is 3.7 mm in diameter, resulting in a laminar jet Reynolds number of  $\approx 230$ . Upon leaving the burner, the premixed gases entrain and mix with ambient air, producing a stable, reproducible flame with no flashback risk.

#### 2.2 Mid-IR laser absorption spectroscopy

A scanned-wavelength direction absorption technique is utilized to spectrally-resolve select rovibrational lines of the target species [14]. For CO<sub>2</sub>, we utilize an interband cascade laser (ICL) with  $\approx 5$  mW output power near 4.19  $\mu$ m to probe the R(0,58) line at 2384.189 cm<sup>-1</sup>, as well as the doublet line pair R(1,105) and R(1,106) at 2384.327 cm<sup>-1</sup> and 2384.331 cm<sup>-1</sup>, respectively, to measure CO<sub>2</sub> mole fraction and vibrational temperature [15]. For CO, we use a quantum cascade laser (QCL) centered near 4.97  $\mu$ m with  $\approx 50$  mW output to probe the P(0,31) and P(1,26) lines at 2008.53 cm<sup>-1</sup> and 2006.78 cm<sup>-1</sup>, to similarly recover mole fraction and

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Fig. 2. (*Left*) Example 200-by-2 pixel region of interest on flame. (*Top*) Background signals ( $I_0$ ) without flame (gray) and absorbance signals ( $I_t$ ) with the flame (color) for different pixels (10 laser scans averaged). (*Bottom*) Example linecenter transmission for each species.

temperature [16]. For C<sub>2</sub>H<sub>6</sub>, we use an ICL near 3.34  $\mu$ m with  $\approx$  1 mW output power to scan over a collection of lines near 2996.9 cm<sup>-1</sup> comprising the <sup>R</sup>Q<sub>3</sub> branch of the  $v_7$  C-H stretch band [17].

The lasers are injection-current scanned across specified wavenumber ( $\nu$ ) ranges at 400 Hz, corresponding to 100 data points per laser scan period on each pixel (at the 40 kHz camera frame rate). The burner is continuously translated vertically during the data acquisition. Example intensity scans for pixels corresponding to different radial locations in the flame are shown in Fig. 2. For each pixel, we performed a running time-average of 10 scans during the vertical translation, resulting in an effective vertical resolution of ~0.125 mm. Stage translation involved 240,000 frames collected in 6 seconds, resulting in a 366 MB data file. The high-speed IR camera (Telops TS-IR-MW) is equipped with a 1 GB buffer memory. For each laser setup, five separate flame measurements (or vertical passes) were taken, increasing the total sample size to 50 scans per vertical interval and facilitating uncertainty analysis.

For a non-uniform gas medium axially-symmetric in r [cm], the Beer-Lambert law integrated over  $\nu$ —or the projected integrated absorbance area  $A_{j,proj}(r)$  [cm<sup>-1</sup>]—is expressed for each line-of-sight or pixel in Eq. 1 [1],

$$A_{j,proj}(r) = \int_{-\infty}^{\infty} \alpha_{\nu} d\nu = \int_{-\infty}^{\infty} -\ln\left(\frac{I_t}{I_0}\right)_{\nu} d\nu = \int_0^{L(r)} K_j(r) dl$$
(1)

where  $\alpha_{\nu}$  is spectral absorbance,  $I_0$  is incident intensity (measured or inferred without the flame), and  $I_t$  is transmitted intensity (measured with the flame). L(r) [cm] is the aggregate path length at radial position r from the axis of symmetry. Individual pixel measurements of  $\alpha_{\nu}$  for each rovibrational transition j of interest, along with associated measurement uncertainties, are shown in Fig. 3. As indicated, 20 to 30 data points are sampled over the spectral domain for each transition. We attain  $A_{j,proj}(r)$  by fitting a Voigt function to the measured  $\alpha_{\nu}$  profile for each line j [14]. Although the projected absorbance lineshapes are not expected to perfectly reflect a Voigt profile, the residuals after fitting proved sufficiently low (~2%) to justify the convenience of this approach for recovering areas.

Generally, the noise level on individual camera pixels after the moderate averaging was similar to that observed using conventional photovoltaic detectors [10], yielding similar precision, but



Fig. 3. Absorbance  $\alpha_{\nu}$ , Voigt fits, and residuals for transitions corresponding to (*Left*) C<sub>2</sub>H<sub>6</sub>, (*Middle*) CO, and (*Right*) CO<sub>2</sub>. Absorbance measurements are averaged from 10 laser scans, and shaded regions indicate uncertainty corresponding to 95% confidence intervals.

uncertainties associated with baseline fitting were larger in some cases. For a signal-to-noise ratio (SNR) criteria of SNR  $\geq$  5, this corresponded to a typical minimum measurable and acceptable  $\alpha_{\nu}$  of ~0.01 for all transitions j. For the interband cascade lasers, a measured baseline signal was used to capture the ambient absorption along the beam path and non-linearity in laser output. However, for the quantum cascade laser (probing CO), temporal fluctuations in intensity scan-to-scan-attributed to unsteady diffraction internal to the camera-precluded the use of a measured empirical baseline  $I_0$  signal, instead requiring a baseline fitting scheme on the measured  $I_t$  that added measurement uncertainty. Various polynomial fits on the non-absorbing regions of the scan were attempted. As can be seen in Fig. 3, the Voigt fit of the P(1,26) line is more strongly influenced by the baseline fitting choice than that of the P(0,31) line, partly owing to its lower  $\alpha_{\gamma}$ . The mean values of a 3rd order polynomial baseline fit yielded more consistent  $A_{i,proj}(r)$  and so were used for all data reported in this study while the baseline fitting variation was incorporated into the measurement uncertainty. For CO<sub>2</sub>, we attain  $A_{j,proj}(r)$  for the R(1,105) + R(1,106) doublet line by employing a sequential Voigt fitting scheme in which a fit of the R(0,58) line is subtracted from the overall measurement as shown in Fig. 3. Notably, ambient CO<sub>2</sub> was measured in the baseline  $I_0$  signal (mostly along the ~80 cm optical path outside of the flame). This background  $CO_2$  absorbance varied little (~1%) between the flame on or off condition and was determined to contribute less than  $\pm 400$  ppm of error to the mole fraction results. Further details on the sequential fitting procedure and ambient CO<sub>2</sub> uncertainty are documented in previous work [10].

Images constructed from measured  $A_{j,proj}(r)$  of selected transitions during burner translation are shown in Fig. 4, highlighting the spatial resolution capability of the technique. These lineof-sight measurements of  $A_{j,proj}(r)$  have a horizontal pixel resolution of 50  $\mu$ m/pixel, which was determined by placing a calibration card of known dimensions in the imaging plane and measuring the number of pixels that reported a blocked transmission intensity of less than 20%. It can be noted that the images demonstrate moderate streak-like discontinuities along the vertical direction, which we attribute to residual diffraction effects. These mild artefacts in the raw data generally do not propagate to the reconstructions of temperature and mole fraction due to the regularization methods employed (to be further discussed). Recalling Eq. 1, the thermochemical properties of interest are embedded in the radially-resolved absorption coefficient  $K_j(r)$  [cm<sup>-2</sup>], given by Eq. 2,

$$K_i(r) = PS_i(T(r))X_{abs}(r)$$
<sup>(2)</sup>

where total pressure P [atm] is assumed constant,  $S_j(T(r))$  [cm<sup>-2</sup>/atm] is the linestrength of transition j at temperature T(r) [K], and  $X_{abs}(r)$  is the mole fraction of the absorbing species.



Fig. 4. (*Far Left*) Photograph of flame with the IR-imaged region outlined. (*Right*) Projected absorbance areas  $A_{j,proj}$  for selected rovibrational transitions of C<sub>2</sub>H<sub>6</sub>, CO, and CO<sub>2</sub>.

#### 2.3 Tomographic reconstruction and thermometry

Assuming the flame is axisymmetric and steady, 1D classical absorption tomography can be applied to each transverse slice of the flame using a numerically integrated Abel transform method, and we can determine radial profiles of  $K_j(r)$  from  $A_{j,proj}(r)$  [1, 10, 13]. To do this, the flame region is divided into equally spaced annular rings and the radial absorption coefficient distribution is approximated by a quadratic function near radius r using the Abel 3-point (ATP) method [18]. The measured  $A_{j,proj}(r)$  (Fig. 5) are smoothed and deconvoluted using Tikhonov regularized Abel inversion to address the inherent ill-conditioned nature of the projection matrix. A suitable regularization parameter is determined from the *L*-curve method following Daun et al. [19] to be  $\lambda \approx 1$  and is used for all reconstructions. Since this process uses information from neighboring pixels, it reduces the effective tomographic resolution [1, 20].

We scan multiple transitions j and thus can determine multiple  $K_j(r)$ , shown in Fig. 5. Then, using established two-line thermometry methods [10, 14], we infer gas temperature T(r) from a



Fig. 5. Data analysis for a row of pixels (z = 2.5 mm). Shaded regions indicate uncertainty. (*Far left*) Path-integrated absorbance area,  $A_{j,proj}(r)$ . (*Left*) Reconstructed absorption coefficients,  $K_j(r)$ . (*Right*) Vibrational temperatures of CO and CO<sub>2</sub>. (*Far right*) Mole fractions of the species, where a uniform temperature of 400 K is assumed to estimate  $X_{C_2H_6}$ .



Fig. 6. Reconstructed temperature [K] (*left*) and mole fraction (*right*) images for CO and  $CO_2$ . Images have been reflected about the axis of symmetry for reader clarity.

ratio of line-strengths  $S_j(T(r))$  (available in the HITRAN database [21] for the lines of interest), and obtain  $X_{abs}(r)$  per Eq. 2. Here, we treat the integrated collection of  $C_2H_6$  transitions as a single line *j* and approximate  $X_{C_2H_6}$  using a constant temperature assumption based on a thermocouple measurement in the core and the pseudo line-list of spectral parameters from Harrison [17]. Although the uncertainties in temperature and mole fraction varied depending on flame location, CO and CO<sub>2</sub> temperatures could be typically determined within ±350 K and ±130 K, respectively, while mole fractions for each species could be typically determined within ±10% and ±6%, respectively. These uncertainties, including those shown in Fig. 5, are calculated using derivations available in our previous work [10].

### 3. Results

The radial profiles depicted in Fig. 5 demonstrate the ability of LAI to spatially resolve mole fraction of fuels, intermediates, and products of combustion, as well as gas temperature in an axisymmetric flame. Generally, the range of scalar values observed are consistent with that expected over the range of equivalence ratios in the partially premixed C<sub>2</sub>H<sub>6</sub>-air flame. Peak temperatures are in the range of 2200–2400 K ( $T_{ad,max} = 2370$  K). Mass flow readings correspond to an inlet  $X_{C_2H_6}$  of 0.114, which agrees well with the tomographic estimate shown in Fig. 5. For CO and CO<sub>2</sub>, vibrational temperature is shown in regions where the ratio of line intensities could reliably be measured (SNR >5). The aforementioned baseline uncertainty and low absorbance of the P(1,26) CO line in many regions of the flame ( $\alpha_v \sim 0.01-0.02$ ) resulted in relatively large uncertainties in CO temperature relative to CO<sub>2</sub> temperature, and potentially a systematic bias yielding somewhat lower CO temperatures. The spatial evolution of these temperatures and species mole fractions can be inspected by assembling the transverse slices along the vertical axis, such as in Fig. 5, to produce two-dimensional reconstructed images of the flame.

Images of temperature and mole fraction from both CO and CO<sub>2</sub> measurements are shown in Fig. 6. For each species, mole fraction and temperature is shown for heights *z* that include radial measurements that extend to the ambient boundary condition (i.e. zero absorption after correcting for ambient concentrations), a requirement for performing 1D classical absorption tomography. This primarily affects CO<sub>2</sub>, which rapidly diffuses out of the available pixel window at z > 20 mm. The temperature is lower in the core of the flame and generally peaks just beyond the radius of the burner, which corresponds to the interface of CO to CO<sub>2</sub> terminal oxidation. As the height above the burner *z* increases, *T* also increases, corresponding with an increase in CO<sub>2</sub> concentration seen in the mole fraction images. CO concentration is largely confined to a

region between the initial oxidation of  $C_2H_6$  and terminal oxidation to  $CO_2$ , providing a good indicator of the reaction zone. At larger *z*, some unsteadiness in the flame becomes more apparent with entrainment of ambient air, precluding reliable reconstruction efforts without an increase in sample size of flame experiments. For the heights imaged, there is little  $CO_2$  diffusion into the core of the flame, though  $CO_2$  monotonically diffuses outward radially as *z* increases.

#### 4. Discussion and conclusions

The aforementioned results demonstrate tomographic laser absorption imaging (LAI) as an effective technique for quantitative spatially-resolved measurements of multiple species and temperature in axisymmetric reacting flows. LAI provides a superior combination of spatial resolution and data collection bandwidth compared to previous laser absorption tomography techniques utilized in the mid-wave infrared. The effective spatial resolution for line-of-sight absorbance is ~50  $\mu$ m in the horizontal direction and ~125  $\mu$ m in the vertical direction, representing approximately an order of magnitude improvement over tomographic techniques for which laser beam size is the limiting factor in spatial resolution [8–12]. Utilizing the LAI setup described in this paper, 2D mapping of an axisymmetric steady flame on the centimeter scale can be completed in less than 10 seconds for a particular laser setup. Compared to tomography techniques utilizing multiple mechanical translation stages on similarly-sized flames, this represents a ~200-fold reduction in data collection time [10]. As such, for quasi-steady flame studies requiring many repeated measurements for statistically significant conclusions or high mass flux experiments that require large amounts of fuel, LAI provides an enabling new method to quantitatively and efficiently characterize fluid-chemistry interactions.

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