Development of Robust Ultrafast CARS Thermometry and Species Detection

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Advancing Renewable Aero-Propulsion

- **Grand challenge:** air-transportation/energy security/combustion
  - Reduced emission of pollutants from aircraft NOx, CO, CO$_2$, UHC, and soot

“Deep insight into multiscale chemical interactions can only be obtained from spectroscopic measurements garnered in spatial and temporal correlation.”

Research activities and areas of impact
Time- and spatially resolved optical diagnostics for combustion analysis

- Challenges: Parameter determination in reacting flows
  - Major- and transient species detection
  - Particulate chemistry
  - Temperature field
  - Mixture fraction
  - Flow field
  - Spatial- and temporal correlation (multiscale analysis)

- Strategy: Snap-shot coherent Raman imagery
  - Simultaneous hyperspectral imaging (x, y, λ) in a single-laser-shot.
  - Benchmarking: Accuracy, Precision, Sensitivity, Resolution and Field-of-view.

CARS imagery in flames:
Time- and spatially resolved optical diagnostics for combustion analysis

• Challenges: Parameter determination in reacting flows
  – Major- and transient species detection
  – Temperature field
  – Spatial- and temporal correlation (multiscale analysis)

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• Objectives: High-fidelity experiments in combustion systems

Experiments informs theory and vice versa

Device validation
- Flameless Combustor

Development of predictive engineering models

TuDelft
Challenge the future
Why should we use CARS?

- Most accurate technique for **thermometry** in reacting flows (wide range of operational conditions).

**Background nanosecond CARS**

- **Actual temperature**
- **Evaluated temperature**
- Inaccuracy ~2-3%
- Single shot precision ~4-5%

**Advanced nanosecond CARS**

- **Actual temperature**
- **Evaluated temperature**
- Inaccuracy ~0%?
- Single shot precision ~4-5%

**Goal!**

- **Improved Accuracy** – Spectroscopic modelling (Raman linewidths, ...)
- **Improved Precision** – Experimental setup (Laser system, ...)
Why should we use CARS?

- Most accurate technique for **thermometry** in reacting flows (wide range of operational conditions).

  - Nanosecond CARS characteristics:
    - Non-intrusive, in-situ probe
    - High temporal resolution (~10 ns)
    - High spatial resolution (~100 µm x 100 µm x 1-2 mm)

- Vibrational CARS, Rotational CARS

Inaccuracy ~2-3%
Single shot precision ~4-5%
Why should we use CARS?

- Most accurate technique for **thermometry** in reacting flows (wide range of operational conditions).

- Nanosecond CARS characteristics:
  - Non-intrusive, in-situ probe
  - High temporal resolution (≈10 ns)
  - High spatial resolution (≈100 µm x 100 µm x 1-2 mm)

- Vibrational CARS, Rotational CARS

- Two-beam femtosecond/picosecond CARS
  - Picosecond temporal resolution (Near collision independent - Raman linewidths)
  - Improved spatial resolution (40 µm x 40 µm x 0.5 mm)
  - 1D and 2D imaging capabilities

Inaccuracy ~2-3%
Single shot precision ~4-5%

Inaccuracy < 2-3%
Single shot precision ≈1%

< 0.5 mm
Two-beam femtosecond/picosecond CARS

Simplified generic phase-matching- and impulsive excitation scheme.

Energy conservation

Laser driven transitions (Q and S)

Phase-matching (momentum conservation)

Vector mismatch
\[ \Delta k = k_{\text{physical}} - k_{\text{geometrical}} > 0 \]

Raman shift

Beam crossing angle (\(\theta\))

All parallel beams

Spectroscopy in the time-domain

Molecular response

fs Stokes

fs pump

ps probe

Time delay / ps
Two-beam femtosecond/picosecond CARS

Air (79% N₂ and 21% O₂) at room temperature

N₂ spectra at two different temperatures

T = 300 K

T = 1700 K
Examples of coherent Raman spectra for some combustion relevant species

- Specific selection rules (transitions)
  - ro-vibrational O-, Q-, S-branch ($\Delta \nu = 1, \Delta J = 0, \pm 2$),
  - pure-rotational O, S-branch ($\Delta J = \pm 2$)
Direct coherent Raman temperature imaging and wideband chemical detection

- Canonical sooting hydrocarbon flat-flame used to benchmark the new techniques.

**Premixed burner principle**

- Fuel + oxidizer
- $\phi = 2.35$
- $HAB=2\text{mm}$
- $T \sim 1750 \text{ K}$
- $HAB=1\text{mm}$
- $T \sim 800 \text{ K}$

**Photo: M. Campbell**

**Burner design (Michelsen group, Sandia)**
**Side wall quenching burner**
- 1D-CARS temperature- and chemical imaging

- **Motivation**
Flame-wall interaction plays a key role in the formation of pollutants in a combustion chamber, such as UHC and CO.
Two-beam 1D-CARS near-wall imaging

- Automatically overlapped pump/Stokes fields, temporally and spatially, makes the technique more robust and higher pulse energy available.
- Spatial sectioning (probe volume):
  \[ \approx 40 \, \mu m \text{ (Beam waist)} \times 40 \, \mu m \text{ (Coherent point-spread function)} \times 0.5 \, mm \text{ (Interaction length)}. \]
Multiparameter spatio-thermochemical probing of flame-wall interactions

- Concurrent detection of $N_2$, $O_2$, $H_2$, (CO), $CO_2$, and $CH_4$ is achieved.
- The excellent imaging resolution allows for thermochemical states of the thermal boundary layer to be probed to within $\sim 40 \ \mu m$ of the interface.
FWI at enhanced turbulence intensities
(Work-in-progress)

- Single-shot spatially dependent statistics of the 1D flame-front gradient / thickness / position become possible (improving heat transfer models)
Single-shot hyperspectral CARS in the gas-phase

Temperature imaging

Wideband chemical imaging

Challenge the future
Simultaneous planar imaging and multiplex spectroscopy in a single-shot

Rotational quantum number $J = 4 \quad 5 \quad 6 \quad 7 \quad 8 \quad 9 \quad 10 \quad 11 \quad 12 \quad 13 \quad 14 \quad 15 \quad 16$

- Tunable spectral dispersion, enabling multispecies detection and probing of a larger 2D field.
- Vector diagram to orientate each location of the spatially resolved measurement.
Dispersive Fourier Transform detection of short pulsed CARS/CSRS signals

CARS (~300 cm\(^{-1}\))

Fast read-out in time

Dispersive medium

1 km single-mode optical fiber

90 ps fwhm

Time (ns)

Intensity / arb.units

N\(_2\)

H\(_2\)

Time / ns
Synchronized ps/ fs laser system for time-resolved non-linear optical spectroscopy/microscopy

- Femtosecond laser (ultrafast amplifier)
  7 mJ/pulse @ ~780-810 nm (~35 fs)

- Picosecond laser (SHBC)
  2.0 mJ/pulse @ 400 nm (~10 ps)

Snap-shot chemiluminescence flexible hyperspectral imagery

Acknowledgement:
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It is equally fun to buy an air-treatment system, as it is to buy a vacuum cleaner.
Distributed auto-ignition combustion modes with reduced NOx emission

 Courtesy of Dr. Arvind Gangoli Rao
Conclusions

• Two-beam femtosecond/picosecond CARS
  - Relevant for 0D, 1D, and 2D temperature measurements in flames when high-fidelity information is needed (inaccuracy <2-3%, precision ~1%)
  - Single-shot quantitative measurements for major species in combustion are within reach (species specific dephasing times, spectroscopy models)

• This ultrafast 1D-CARS technique has been successfully employed at:
  1. Flame-wall interaction burner (head-on and side-wall quenching)
  2. Sooty flames provided on a McKenna burner

• Can this advanced laser diagnostics technique be employed for measurements in engines?
  - Technical challenges for the stability of operation (facility temperature and humidity control, propagating TL-beams through optical ports)
Thank you for your attention!