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Development of simultaneous hyperspectral coherent Raman imaging for advancing reduced emission combustion technology

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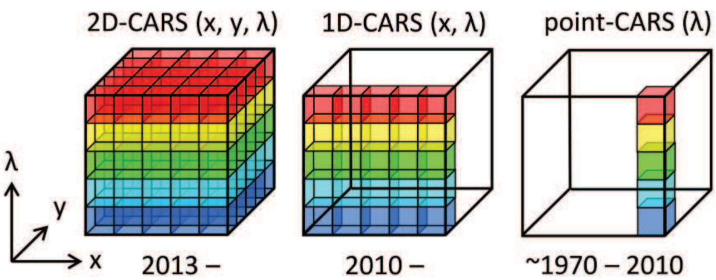
Overall aim and key objectives

Advances in optical imaging techniques over the past decades have revolutionized our ability to study chemically reactive flows encountered in air-breathing combustion systems. Emerging technology for unravelling clean- and efficient heat release is needed for advancing new reduced emission technology, and is on the central agenda for a wide variety of energy production- and transport industry. Combustion of fossil fuels remains our largest source of energy production in the world, and global concerns regarding energy security, environmental pollution, and anthropogenic climate change have motivated a large body of research devoted to the experimental measurement and numerical simulation of combustion systems. Clean combustion engineering is the search for improved efficiency by means of strengthen the systems fuel-economy and lowering the emission of NOx, particulates, CO and unburned hydrocarbons (incomplete combustion). New reduced emission technology, greatly rely upon the ability to control the heat release and the exhaust produced by the exothermic reactions between the fuel and the oxidizer in the chemically reactive flow. For the engineering system design, it exist a significant need to inform on the flame-physics involved based on direct observation of the combustion reaction progress and interaction, which is a demanding task for any measurement technique. Chemically reactive flows are inherently multiscale, fully characterized in three-dimensional space and evolving on rapid time-scales. The combustion environment imposes a significant challenge for diagnostics, where it needs to be collected complete information ideally with correlated-field multi-parameter measurement capabilities, exhibiting high spatial and temporal resolution and provided within a snap-shot to freeze the fast dynamics involved. Concurrent detection of major- and minor molecular species (multiplexing) and determining the three most important scalars; the temperature, the flow-field, and the mixture fraction, is vitally important in studies of the reactive flow. The temperature marks the evolution of heat release and energy transfer, while species concentration gradients provide critical information on mixing and chemical reaction.

Optical imaging techniques have the advantage of being non-invasive, which means that the studied process is not significantly perturbed by the measurement technique, and allowing for the acquisition of statistics *in-situ*. Spectroscopy offer intrinsic chemical specificity, in that different classes of molecules have specific spectral signatures serving as unique fingerprints for their identification. Laser-based diagnostics may in general provide measurements with exceptionally high spatial- and temporal resolution, which is important in producing reliable and accurate experimental data. Coherent anti-Stokes Raman spectroscopy (CARS) is one such versatile technique, which has had a profound impact on a wide variety of fields. It was pioneered in composition- and temperature measurements almost 40 years ago, and is referred to as authoritative with the level of accuracy and precision it may provide. A limitation still, has been its main applicability as a single point-measurement technique, where the experimenter needs to raster-scan the measurement samples assembling the spatial image. Because many complex systems can be fully characterized in multidimensional space, there is a large motivation for the advancement of multidimensional CARS imaging techniques.

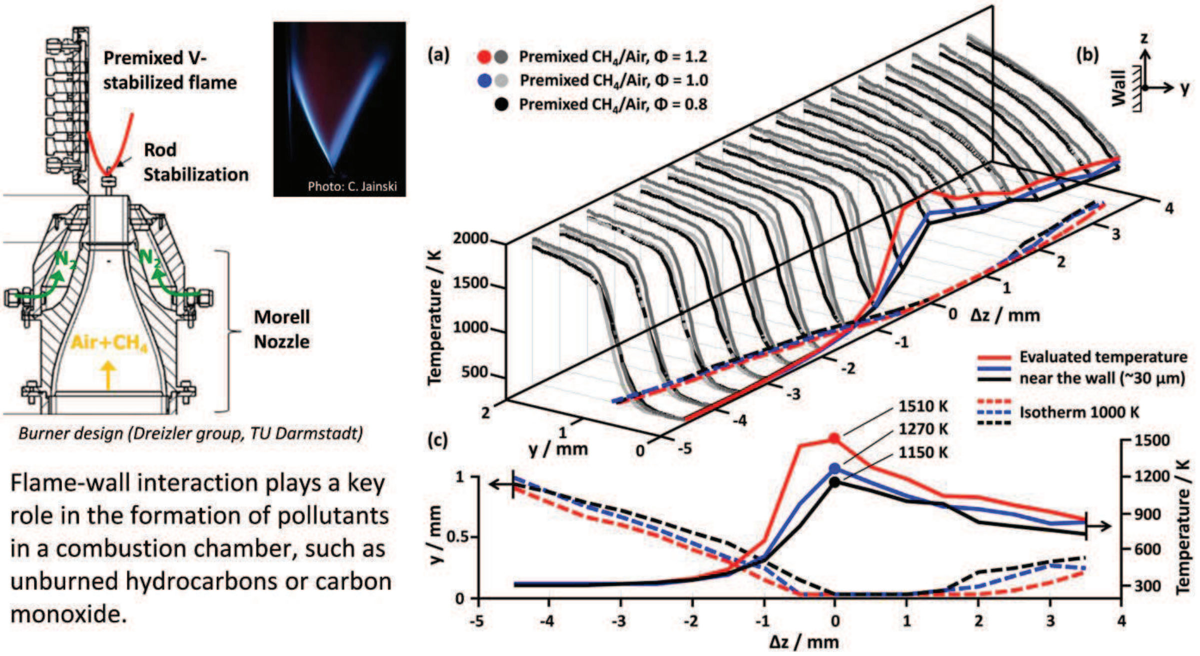


The history development of gas-phase simultaneous CARS imaging. The first 2D-CARS imaging technique was demonstrated in 2013, by me and Sandia researcher Christopher J. Kliewer. The hyperspectral 3D data cube (λ,y,x) is retrieved in a snap-shot (non-scanning) procedure.

CARS imaging of ignition-kernels and flame propagation in vitiated flows

Flameless Combustion (FC) has emerged as one interesting option to be investigated for future gas-turbine operation, inspired by the progress obtained in furnaces- and fluidized-bed combustion applications (see for example [1]). FC is characterized by fuel oxidation at lower peak temperatures and with non-visible flames for the naked eye, where the combustion process occur in a more distributed reaction zone. The FC mode has demonstrated low emissions of NOx, particulates, CO and unburned hydrocarbons, and would be very beneficial for gas-turbines because of the more uniform temperature profile leading to lower combustor pattern factor, and reduced perturbation from thermo-acoustics in comparison with conventional combustors. To implement the FC mode in aero-engines and gas-turbines used for power production is not straightforward, however; the implications related to the different physical scales relative a combustion furnace, makes it a difficult technical challenge and more insight is needed to mature this promising technology. The gas-turbine combustors need to be contained in a restricted volume, and requiring a broad operational range obtained with high stability and with low overall pressure losses.

We will carry out the realization experiment of this new combustion mode in a fuel-flexible FC combustor (~250kW), with access to compressed air provided at a maximum mass-flow rate of ~250g/s up to 5 bar. The combustor prototype will be operated with different fuels (methane and/or hydrogen) in mixtures with air and dilution gases (N_2 and CO_2) at variable mass-flow rates and thermal inputs. The air and dilution gases are pre-heated prior to the injection with the utility of exploring a range of input temperatures using an electric heater (maximum power of ~50 kW, ~900K). The flexibility will enable the attainment of FC and to acquire quantitative data on this combustion mode with its boundaries. The examination provided by the current multi-parameter diagnostics platform is unique, since the *in-situ* information on temperature and its fluctuations is particularly important for understanding the thermal NO formation process, which is the predominant one in gas-turbine combustion with a residence time ranging from 10 to 30 ms. CARS multiplexing across the entrance to the exit section of the combustor, will be able to provide information on the complete oxidation of the reactants, the air-entrainment- and the exhaust-gas-recirculation processes as a consequence of the complex fluid-dynamics involved. The realization experiment will serve as the primary input developing computational codes (CFD) on FC, which will be performed in direct connection to the experiment. The CARS system is built from the <35 fs output of a high-power femtosecond regenerative amplifier with a pulse energy of ~7mJ provided at a 1kHz repetition-rate. The setup is implemented with the two-beam femtosecond/picosecond phase-matching scheme, recently utilized for multi-parameter spatio-thermochemical probing of flame-wall interactions [2] (displayed in the figure below), with an efficient coherent excitation bandwidth spanning the pure-rotational manifold of all Raman active species. The narrowband ps-duration probe-beam centered at 400nm, is efficiently produced by the principles of second-harmonic bandwidth compression [3], originating from a portion of the fs laser output being repetition-wise synchronized with the fs pump/Stokes-beam, and automatically phase-locked to the impulsive excitation pulse at the experiment with an arbitrary time-arrival. This ultrafast coherent Raman imaging is offering a well-benchmarked ~1% relative standard-deviation in single-shot precision thermometry. The new utility of image post-processing available from the spatially correlated measurements will improve the precision of the flame-data even further. In this way, the direct imaging will bring new possibilities for noise reduction with robust performance, and the claimed stability of the FC mode which is still an open question, can now uniquely be assessed at each the measurement locations in the combustor chamber. Statistics on single ignition-kernels is a goal and to watch the flame propagation from these events.



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