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Development of two-beam femtosecond/picosecond CARS for high-fidelity thermometry in flames

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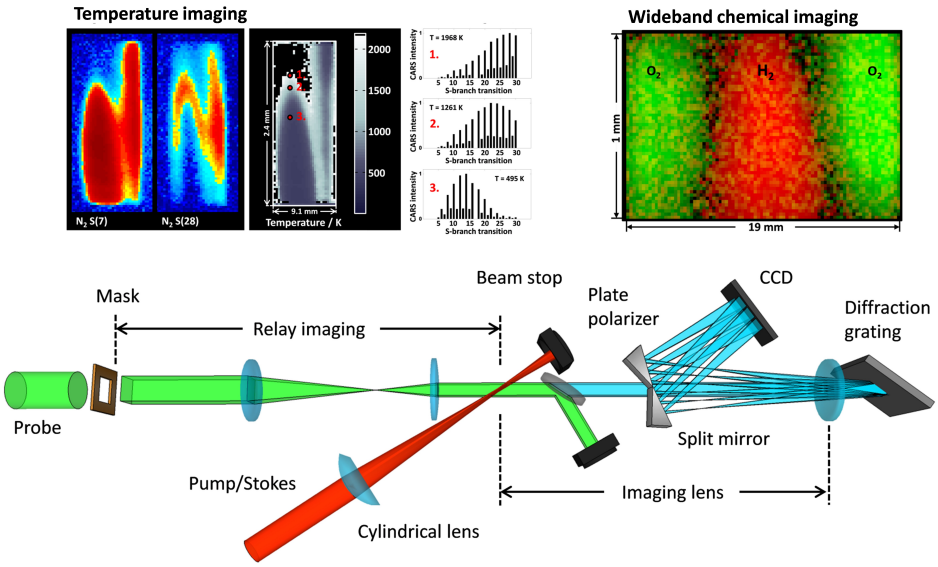
Abstract

We develop simultaneous ultrafast CARS thermometry and species detection as a unique tool to investigate combustion modes with reduced emissions of NOx, particulates, CO and unburned hydrocarbons. Coherent anti-Stokes Raman spectroscopy (CARS) has been developed over the last 4 decades for various gas-phase-, plasma-, flame- and combustion applications and is referred to as the “gold standard” for nonintrusive spectroscopic measurements in harsh environments. It is the combination of excellent chemical selectiveness, i.e. the ability in targeting distinct quantum states of individual molecular species, and the coherent properties of the generated signal beam, which makes this technique very powerful. For most of the gas-phase applications, with rapidly changing events ranging from μs to ms, there is a constraint in performing instantaneous measurements and is beneficially fulfilled by the inherent property of the Raman process. It is the time-duration of the probe pulse which ultimately limits the temporal resolution, where today existing techniques, implemented with ultrafast laser systems operate around or even below collision lifetimes. Still, while a reacting flow is fully characterized in three-dimensional space, a limitation of CARS has been its main applicability as a point-wise measurement technique. Establishing ultrafast CARS for single-shot imaging, however, requires designing robust schemes for both the signal generation and the signal detection. Because the physical scalars quantifying an event in a reacting flow exist only for an instantaneous moment of time, laser-scanning or sample-rastering procedures may not be employed. Here we discuss two-beam femtosecond/picosecond CARS relevant for 0D, 1D, and 2D temperature measurements in flames, and we show that single-shot quantitative measurements for major species in combustion are within reach.

Keywords: Atomic and molecular physics; Nonlinear optics; Ultrafast optics; Laser diagnostics



Single-shot hyperspectral CARS in the gas-phase