DEVELOPMENT OF ULTRAFAST SPECTROSCOPY IMAGING TO STUDY HYPERSONIC FLOW PHENOMENA

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Abstract

Progress has been made on the development of measurement capabilities for the study of carbon monoxide (CO) and carbon dioxide (CO_2) energy distributions post-shock near a reentry vehicle surface. Hybrid femtosecond/picosecond (fs/ps) coherent anti-Stokes Raman scattering (CARS) models for CO and CO_2 have been developed and model predictions are compared against observed spectra taken within a near-adiabatic laboratory flame. Simultaneous CARS thermometry of CO and N_2 as well as CO_2 and O_2 are performed for model validation. Excellent agreement between CO and N_2 thermometry results are observed. A temperature discrepancy between the best-fit model temperatures of CO_2 and O_2 are observed and possible causes for this discrepancy are discussed.

Introduction

During hypersonic flight and atmospheric entry, high vehicle speeds result in a strong shock and shock-induced plasma, which can cause heating and ablation of the vehicle surface. This challenge can be addressed through improved understanding and characterization of the reactive gas environment near the vehicle. The interaction between the surface of a vehicle and hypersonic flow, while critically important to the design of robust and reusable vehicle materials, is not well-understood.[1] In order to improve the utility of ground-based testing and simulation, the thermal properties and chemistry of the reacting flow must be accurately characterized. Heat shield materials for reentry vehicles are often tested in arc-jet facilities which provide realistic flow enthalpies and velocities, but highly dissociate the flow prior to the test article. This has led to the questioning of the representativeness of test results from such facilities.[2]

In atmospheres that are primarily composed of carbon dioxide, such as that of Mars and Venus, the species responsible for entry vehicle surface reactions are atomic oxygen (O) and carbon monoxide (CO).[3] Improving the understanding of gas and surface reactions involving O, CO, and vehicle materials at hypersonic conditions is necessary to improve materials and overall vehicle design. These species are also key combustor intermediates and products in air-breathing scramjets operating at atmospheric conditions on Earth. Thus, thermometry measurements of CO and CO_2 have broader applications beyond the primary objective of understanding flow phenomena near the surface of a reentry vehicle.

Optical-based measurements such as coherent anti-Stokes Raman scattering (CARS) spectroscopy are advantageous for reacting flow characterization because they are generally nonintrusive. In other words, they can be used to obtain quantitative thermodynamic information about the flowfield without disturbing the flow or interrupting reactions. CARS can be used for simultaneous probing of temperature and species distributions and has recently been employed for thermometry of carbon dioxide.[4]

CARS is a laser-based measurement technique that employs the nonlinear interaction of three beams (pump, Stokes, and probe) in a test gas to produce a coherent laser-like CARS beam containing information about the molecular energy distributions and species concentrations within the gas at the probe location. In hybrid femtosecond/picosecond (fs/ps) CARS spectroscopy, ultrashort <100 fs laser pulses are used to excite a Raman coherence, while a picosecond pulse is used to probe the spectral response. This method allows a measurement period duration similar to the timescale of molecular collisions, so that the measurement is minimally affected by such collisions. If the three laser beams are focused at the CARS probe volume, spatial resolution on the order of hundreds of microns can be readily achieved.[5]

Methods

CARS model

Development of a time-domain hybrid fs/ps CARS model for CO_2 has, to the author's knowledge, only

been presented in one previous study[4] and thus comprised a significant portion of this project. To generate synthetic CARS spectra of a molecule, information about that molecule's rotational and vibrational energy levels, Raman transition intensities, and collisionally-broadened transitions linewidths must be known. For diatomic molecules with a single vibrational degree of freedom such as N_2, O_2, and CO, these molecular properties are well-understood and can be accurately estimated with relatively simple equations.

Carbon dioxide, on the other hand, possesses three atoms and four vibrational degrees of freedom which significantly complicate the analysis. Rovibrational energy levels can be estimated with reasonable accuracy using an anharmonic force constant model of CO_2[6] and the Raman transition intensities can be estimated with an empirical relationship.[7,8] Collisionally-broadened linewidths can be modeled using a simple polynomial model fitted to experimental linewidth data or a more complete energy-corrected sudden (ECS) model, which allows for the effects of linemixing (collisional narrowing) to be included.[9,10] In this work, constants for the lowest ~200 vibrational states of CO_2 are taken from literature,[11] while constants for higher vibrational states, which are necessary for the calculation of hot CO_2 spectra, are estimated using the anharmonic force constant model. The polynomial linewidth model was used here to reduce the computational time required for spectral synthesis. The models for CO and CO_2 are described in more detail in a paper that was presented at the 2021 AIAA SciTech Conference.[12]

Laboratory flame

To produce high-temperature CO and CO_2 for detection and thermometry with the CARS system, a near-adiabatic H_2-air flame provided by a Hencken burner was used. Since CO and CO_2 do not naturally occur in large quantities in an H_2-air flame, either CO or CO_2 was seeded into the air inlet of the burner depending on the molecule of interest. In general, the adiabatic flame temperature increases with increasing CO reactant mole fraction, while the opposite is true for CO_2. Thus, the flame temperature could be varied by adjusting the flow rate of CO or CO_2. Adjustment of the H_2-air equivalence ratio (ϕ) provided additional control of the flame temperature. Thus, test conditions spanning a range of temperatures with sufficient CO or CO_2 mole fraction for CARS detection could be produced by varying equivalence ratio and CO/CO_2 flow rate simultaneously.

The CARS measurement volume was positioned 30 mm above the burner surface, where the flame should not have any significant temperature gradients, and the flame temperature should be approximately equal to the adiabatic flame temperature.[13] At all test conditions, the air flow rate was kept at a constant 55 SLPM, and the N_2 coflow was held at either 85 or 100 SLPM. Flow rates of CO and CO_2 were varied between 3 and 25 SLPM across the range of test conditions.

Hot CO was probed with CARS while diluting the air inlet to the burner with CO, while hot CO_2 was probed while diluting with CO_2. While the NASA CEA (Chemical Equilibrium with Applications) code can be used to calculate the adiabatic flame temperature of each test condition, it is likely that the flame was not operating at the adiabatic flame temperature when significant flow rates of CO or CO_2 were used. To ensure nominal operation of the Hencken burner and the laser system, CARS spectra of N_2 were collected across a range of equivalence ratios (ϕ = 0.6, 0.8, 1.0, and 1.2) without CO or CO_2 seeding.

CARS system setup

Ultrashort 55 fs, 800 nm pulses at a repetition rate of 1 kHz are produced using a single laser source (Coherent, Astrella) and split to form the pump, Stokes, and probe pulses. A diagram of this system is shown in Figure 1. Part of the beam is sent to an optical parametric amplifier (Coherent, TOPAS), which generates light at user-defined wavelengths depending on the molecule to be studied (e.g. 674 nm for N_2 or 711 nm for O_2), forming the 55 fs pump pulse. The narrowband (~4 cm^{-1}) 800 nm picosecond-duration probe pulse is formed using a $4f$ pulse shaper. The delay of the probe pulse relative to the pump and Stokes pulses was kept fixed at 4.05 ps. The 800 nm Stokes pulses are 55 fs in duration. The per-pulse energies here were approximately for 35 μJ for each pump, Stokes, and probe pulse. A folded BOXCARS phase-matching con-

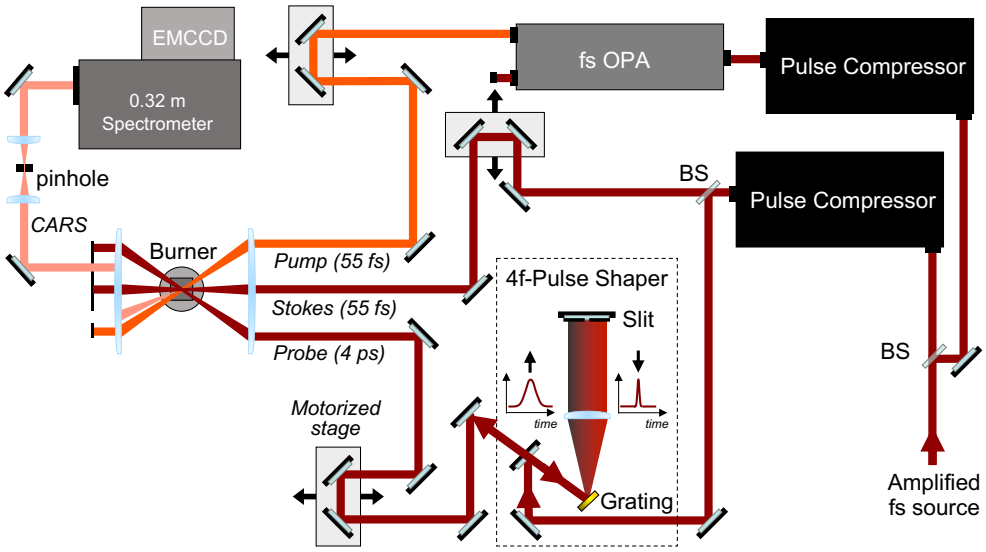

Figure 1: Hybrid fs/ps CARS experimental setup.

figuration with 2.4° crossing angle was used, with 300 mm focusing and collimating lenses. The collimated CARS signal was passed through a spatial filter comprising a 150 mm lens, a 100 μm pinhole placed at the focus of the 150 mm lens, followed by a 200 mm lens to collimate the CARS signal. Finally, the CARS signal was coupled into a spectrometer (Princeton Instruments, SCT-320) and detected with an electron-multiplied CCD (Princeton Instruments, ProEM: 1600 eXcelon 3). Additional laser system details are provided in the AIAA SciTech Conference paper.[12]

The flame temperature at each test condition must be known to validate the CO and CO_2 CARS models, so the temperature was measured using simultaneous CARS measurements of O_2 (when probing CO_2) or N_2 (when probing CO). This is facilitated by the pump/Stokes excitation bandwidth ($\sim$250 cm^{-1}) and the spectral position of the Q-branch of each molecule: O_2 at 1560 cm^{-1} is near CO_2 at 1286 and 1388 cm^{-1}, while N_2 at 2330 cm^{-1} is near CO at 2145 cm^{-1}.

Results

Simultaneously-acquired CARS spectra of CO and N_2 as well as CO_2 and O_2 taken within the Hencken burner flame with corresponding spectra predicted by the CARS model are shown in Figure 2. For the case shown in Fig. 2a, the modeled spectra of both CO and N_2, both at 2700 K, agree well with the experimental spectra for both species. For the case shown in Fig. 2b, the modeled spectrum of CO_2 at 1200 K agrees well with the experimental spectrum. However, the modeled O_2 spectrum at the same temperature appears colder than the observed O_2 spectrum, which is best fit by a 1500 K spectrum. Thus, it is likely that this flame condition was close to 1500 K rather than 1200 K. The 300 K discrepancy between the best-fit CO_2 and O_2 modeled spectra requires further investigation. One possible explanation for this discrepancy is that linemixing was neglected in the CO_2 spectral model, which could cause the model spectrum to appear hotter than an experimental spectrum of a CO_2 at the same temperature.[8]

Conclusion

Recently-developed time-domain fs/ps CARS models of CO and CO_2 have been demonstrated and compared against experimental CARS spectra taken within a laboratory flame. Excellent agreement between the CO model and experiment are observed. A temperature discrepancy between the best-fit model CO_2 spectrum and the flame temperature measured via O_2 CARS thermometry is also observed, and could be due to the lack of linemixing included in the CARS model for CO_2. Future work will include the implementation of a

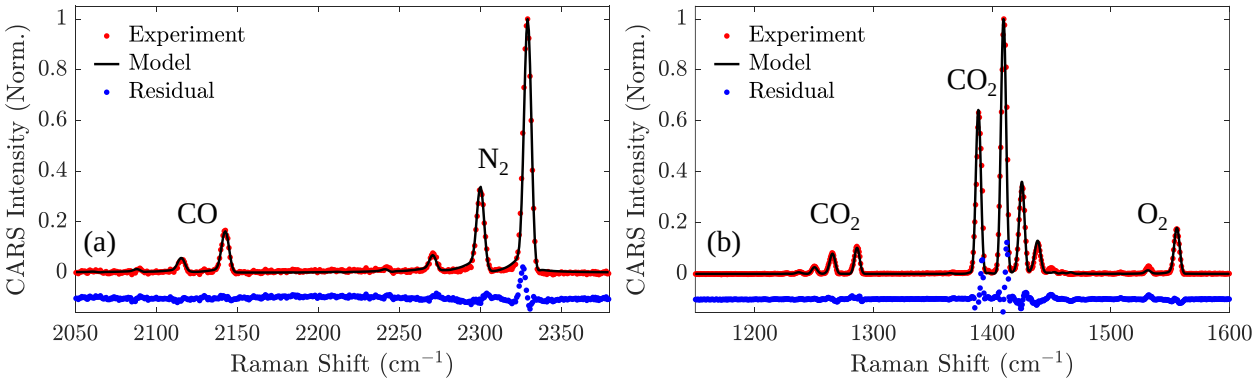

Figure 2: (a) Experimental spectrum of CO and N_2 in a flame operating at $\phi = 0.87$ with 20 SLPM of CO addition, and 2700 K model spectrum overlaid. (b) Experimental spectrum of CO_2 and O_2 in a flame operating at $\phi = 0.6$ with 9 SLPM of CO_2 addition, and 1200 K model spectrum overlaid.

more accurate linewidth model and linemixing for CO_2. Additionally, CARS measurements of CO and CO_2 within a benchtop plasma for model validation at rotational-vibrational nonequilibrium conditions, such as those found in the wake of a shock wave, are planned for the near future.

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