

Femtosecond Coherent Anti-Stokes Raman Spectroscopy in a Cold-Flow Hypersonic Wind Tunnel for Simultaneous Pressure and Temperature Measurements

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Measurements of gas-phase pressure and temperature in hypersonic flows are important to understanding fluid–structure interactions on vehicle surfaces, and to develop compressible flow turbulence models. To achieve this measurement capability, femtosecond coherent anti-Stokes Raman scattering (fs CARS) is applied at Sandia National Laboratories’ hypersonic wind tunnel. After excitation of rotational Raman transitions by a broadband femtosecond laser pulse, two probe pulses are used: one at an early time where the collisional environment has largely not affected the Raman coherence, and another at a later time after the collisional environment has led to significant J-dependent dephasing of the Raman coherence. CARS spectra from the early probe are fit for temperature, while the later CARS spectra are fit for pressure. Challenges related to implementing fs CARS in cold-flow hypersonic facilities are discussed. Excessive fs pump energy can lead to flow perturbations. The output of a second-harmonic bandwidth compressor (SHBC) is spectrally filtered using a volume Bragg grating to provide the narrowband ps probe pulses and enable single-shot CARS measurements at 1 kHz. Measurements are demonstrated at temperatures and pressures relevant to cold-flow hypersonic wind tunnels in a low-pressure cryostat with an initial demonstration in the hypersonic wind tunnel.

I. Introduction

Fluctuations in hypersonic flows around a flight vehicle can cause loading and deformation on vehicle surfaces and have been studied as fluid–structure interactions [1–8]. To better understand these interactions, measurements of gas-phase pressure fluctuations are critical to complement surface pressure and structural deformation measurements. Additionally, simultaneous measurements of pressure and temperature will provide new insight into compressible flow turbulence. A few methods for performing single-shot, non-invasive, spatially resolved pressure or density measurements have been developed including focused laser differential interferometry (FLDI) [9], nanosecond coherent anti-Stokes Raman spectroscopy (ns CARS) [10–12], and femtosecond (fs) CARS [13–15]. Other techniques such as laser induced fluorescence and filtered Rayleigh scattering have been used to perform planar pressure, temperature and velocity measurements but require scanning the laser wavelength and cannot be performed on a single laser shot [16,17]. This work focuses on the development and demonstration of a fs CARS temperature and pressure diagnostic capability for use in Sandia National Laboratories’ hypersonic wind tunnel, where freestream temperatures are typically 35-60 K and freestream pressures are below 1 kPa. [18]. Application of fs CARS in this environment has several challenges including the following.

1. Low gas densities (approximately 5% of atmospheric density) in the hypersonic flow.

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- a. This leads to low CARS signal levels as the CARS signal strength scales with gas density squared. The expected CARS signal level in the hypersonic wind tunnel is 0.25% that of a CARS signal recorded at ambient conditions.
 - b. The low gas density also leads to long Raman dephasing times and the need for relatively long (~ 10 ns) optical delay lines for CARS-based pressure measurements. These long delays are challenging due to spatial constraints near the large wind tunnel facility and the need to maintain spatial overlap of the CARS beams at various delays.
2. Cryogenic gas temperatures of approximately 50 K in the hypersonic flow. At this temperature, all populated rotational Raman lines are spectrally close to the probe laser line, and the gas temperature is based on the relative intensities of only a few (~ 8) rotational lines. The pressure measurement is based on the relative decays of these same few rotational lines at long probe delays.
 3. Implementation of a complex optical setup in a large facility with limited run times.

This paper focuses on addressing these challenges and presents a methodology to successfully perform simultaneous temperature and pressure measurements using fs CARS in cold-flow hypersonic wind tunnels.

II. Experiment Setup

In fs CARS, a broadband fs laser pulse generates a Raman coherence; both pump and Stokes photons may come from a single broadband pulse. The 100-fs pulses used here had enough bandwidth to efficiently excite rotational transitions in the pure-nitrogen wind tunnel flow. After initial excitation, the Raman coherence undergoes dephasing due to gas-phase collisions. Higher gas densities lead to faster dephasing rates. Different rotational Raman transitions decay at different rates, with lower J transitions decaying faster. With ns lasers, pressure measurements can be performed by measuring the linewidths evident in the CARS spectrum [12]. With fs lasers, pressure measurements are performed by quantifying the J -dependent dephasing by using a relatively large probe pulse delay [13]. One advantage of a fs CARS instrument is that pressure-insensitive thermometry can be performed at early probe delays when the Raman coherence has not experienced significant dephasing. In order to extract quantitative pressure data, the gas temperature needs to be known and this is accomplished by using two probe pulses, one at an early delay to measure the gas temperature, and one at a later delay where significant dephasing has occurred to measure gas pressure. This pressure measurement technique has been demonstrated at bench-top conditions [13,14]. The fs CARS technique was chosen for this work as the sensitivity of the diagnostic can be tuned by changing probe time delays [13], and the enhanced data rate available with fs lasers (1 kHz) compared to ns laser systems (10 Hz). Gas thermometry in a hypersonic wind tunnel has been performed previously with a fs CARS instrument [19,20].

For the work presented here, point CARS measurements were performed in two environments: a low-pressure cryostat used to replicate the temperature and pressure of a cold-flow hypersonic wind tunnel, and the hypersonic wind tunnel at Sandia National Laboratories in Albuquerque, New Mexico, USA. The laser system included a regeneratively amplified femtosecond (fs) laser (Solstice, Spectra Physics) producing a 1-kHz pulse train with a pulse duration of ~ 100 fs at 800 nm and a pulse energy of 2.5 mJ. This radiation was split to form the pump/Stokes laser pulse and to pump a second-harmonic bandwidth compressor (SHBC) which produced the probe pulse at 400nm with a pulse duration of 6 picoseconds (ps) and a pulse energy of 400 μ J. The SHBC output was split with $\sim 10\%$ of the energy going to the early probe pulse, and the remainder forming the late probe pulse. Independent time-delay stages were used to control the relative timing of the two ps probe pulses with respect to the fs pump/Stokes pulse. The early and late probe time delays were 4 ps and 5–11 ns respectively. An annotated photograph and simplified diagram of the laser system are shown in Fig. 1.

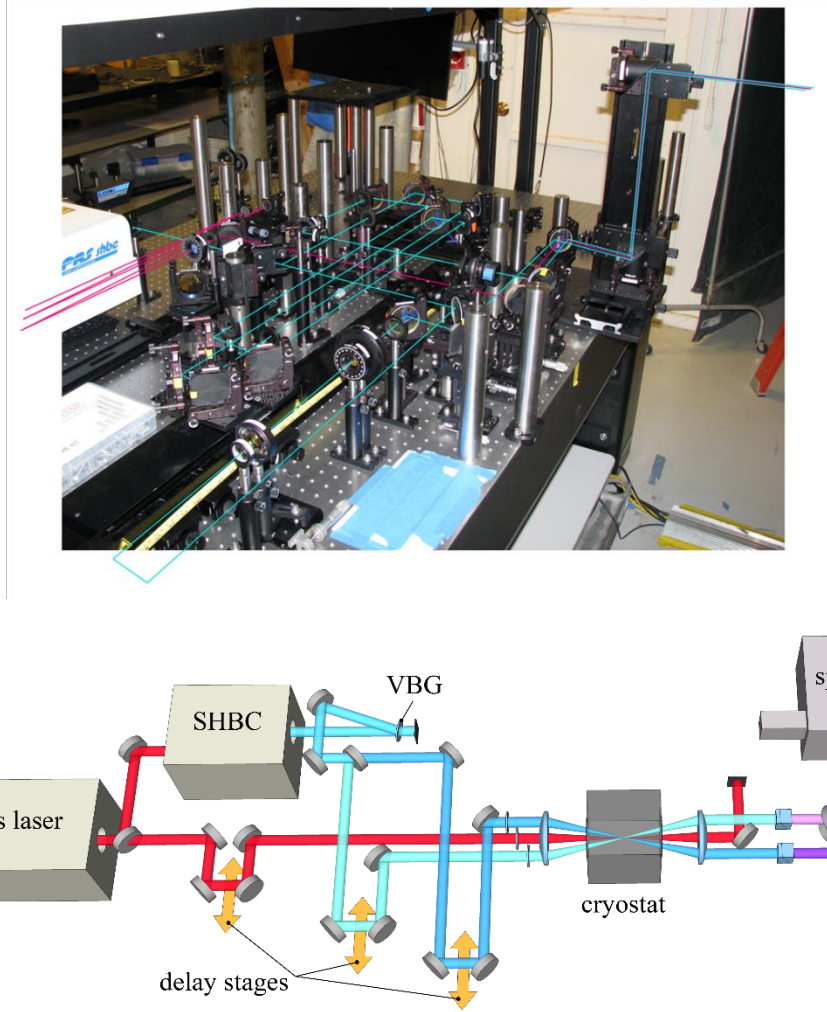


Fig. 1. Annotated photograph of the laser system used to perform measurements in Sandia National Laboratories' hypersonic wind tunnel (top) and simplified diagram of the same instrument used to perform measurements in a low-pressure cryostat (bottom). SHBC – second harmonic bandwidth compressor. VBG – volume Bragg grating.

The fs pump/Stokes pulse and the two probe pulses were directed to the hypersonic wind tunnel test section and overlapped in a planar configuration with the fs pump in the center and the early and late probes on either side as shown in the diagram in Fig. 1. This arrangement is a two-channel version of a two-beam fs-ps rotational CARS technique reported earlier for gas thermometry [21]. The measurement volume length was found to be 2.5 mm long in the direction of beam propagation, and approximately 50 μm in the transverse directions. The final turning mirror and focusing lens were mounted on a translation stage to scan the CARS measurement volume to different regions in the wind tunnel test section. The wind tunnel was run at Mach 8 with pure nitrogen and stagnation conditions of approximately $p_0 = 5.3 \text{ MPa}$ and $T_0 = 595 \text{ K}$. Assuming isentropic expansion to Mach 8, the free-stream conditions in the test section are estimated to be $p_1 = 540 \text{ Pa}$ and $T_1 = 45 \text{ K}$. Immediately after a conical shock from a seven-degree sharp cone model, the conditions are estimated to be $p_2 = 1.2 \text{ kPa}$ and $T_2 = 55 \text{ K}$. These cryogenic temperatures are a unique measurement environment for fs CARS measurements.

The low-pressure cryostat was used as a surrogate environment for technique development and refinement. A photograph of the cryostat is shown in Fig. 2. The cryostat pressure was set using a vacuum pump and a needle valve with a supply of N_2 ; the cryostat pressure was monitored using a convection gauge. The temperature of the cryostat was set using a flow of liquid N_2 to cool the gases in the device, and the internal gas temperature was monitored using a thermocouple placed near the CARS measurement volume.

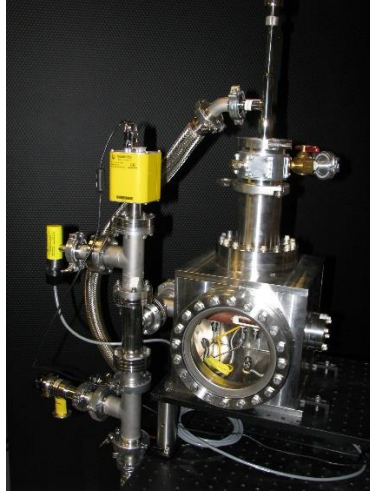


Fig. 2. Photograph of the low-pressure cryostat used in this work to simulate the temperature and pressure found in the hypersonic wind tunnel.

III. Results and Analysis

Sample fs CARS spectra recorded in the hypersonic wind tunnel at $Ma = 8$ are shown in Fig. 2 along with best-fit theoretical spectra. High CARS signal levels were observed for measurements performed in the freestream and after the oblique shock (not shown here).

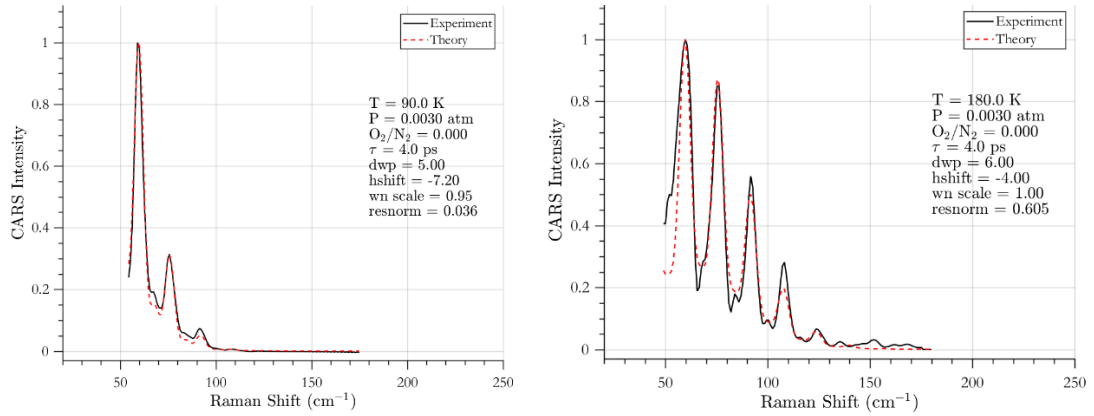


Fig. 3. Sample fs CARS spectra recorded in the free stream of a $Ma = 8$ hypersonic wind tunnel run along with best-fit theoretical spectra.

One of the main challenges encountered was the balance between using enough pump/Stokes laser energy to achieve usable signal levels, but avoiding intrusive perturbations to the flow. For many of the measurements performed to date, the laser energies used proved to be too high which led to unrealistic best-fit temperatures as shown in the plot on the right-hand side of Fig. 2 where the best-fit temperature of 180 K is not realistic for this cold-flow facility. These results led to a study in the low-pressure cryostat on the effect of pump pulse energy on fs CARS thermometry at conditions relevant to a cold flow hypersonic facility.

CARS spectra were recorded in the cryostat with a measured gas temperature and pressure of 104 K and 5 Torr, respectively. CARS spectra were recorded with pump energies of 51–390 μJ and are shown in Fig. 4. The CARS spectra were normalized based on the amplitude of the spectral peak near a Raman shift of 74 cm^{-1} . For pump energies

above about 100 μJ , the pump beam was observed to significantly perturb the gases, leading to a CARS spectrum with higher intensities at larger Raman shifts than the unperturbed measurements. Based on these results, it was determined that a maximum pump energy of 100 μJ should be used in the current experimental setup for measurements in the hypersonic wind tunnel.

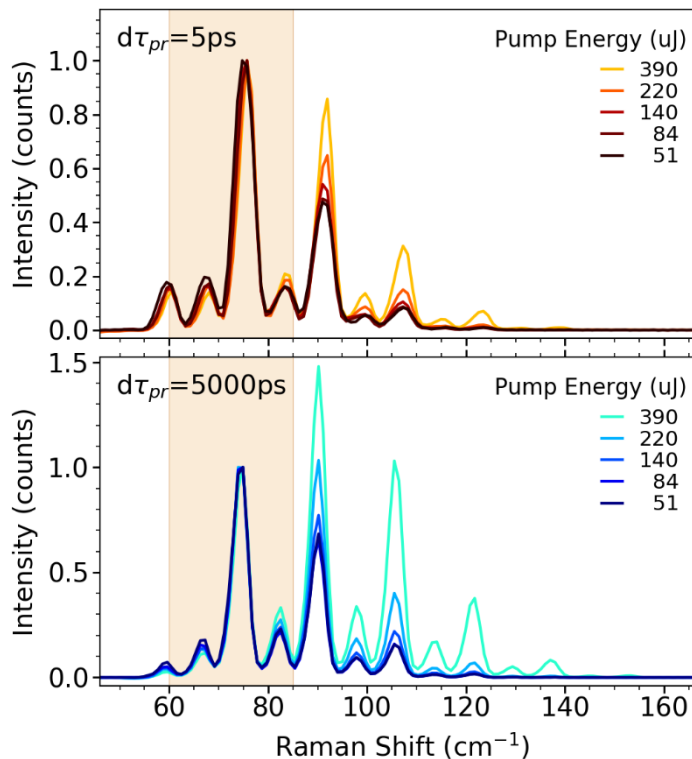


Fig. 4. Sample fs CARS spectra recorded in the cryostat with pure N_2 at $T = 104 \text{ K}$, $p = 5 \text{ Torr}$, with various pump pulse energies.

The use of the SHBC to produce a ps-duration, spectrally narrow probe pulses allowed measurements to be performed at the laser repetition rate of 1 kHz [22,23]. As discussed above, the output of the SHBC was split to form the early and late probe pulses using a polarizing beam splitter. The 400-nm SHBC beam had some significant energy in the spectral wings. Some of this probe light was transmitted through the analyzer polarizer and an angle-tunable short-pass filter, and recorded simultaneously with the CARS signal. This was problematic for measurements in the hypersonic wind tunnel at conditions with low signal levels such as low pump energies and large probe delays. In some cases, the wings of the probe spectrum obscured the CARS signal. To overcome this, a volume Bragg grating was installed near the exit of the SHBC, and the diffracted light from the grating was used as the probe beam. An annotated photograph of this setup is shown in Fig. 5. The volume Bragg grating diffracted a portion of the incident beam from the SHBC. Other CARS instruments have been built with various methods of filtering the output of an SHBC [24–26].

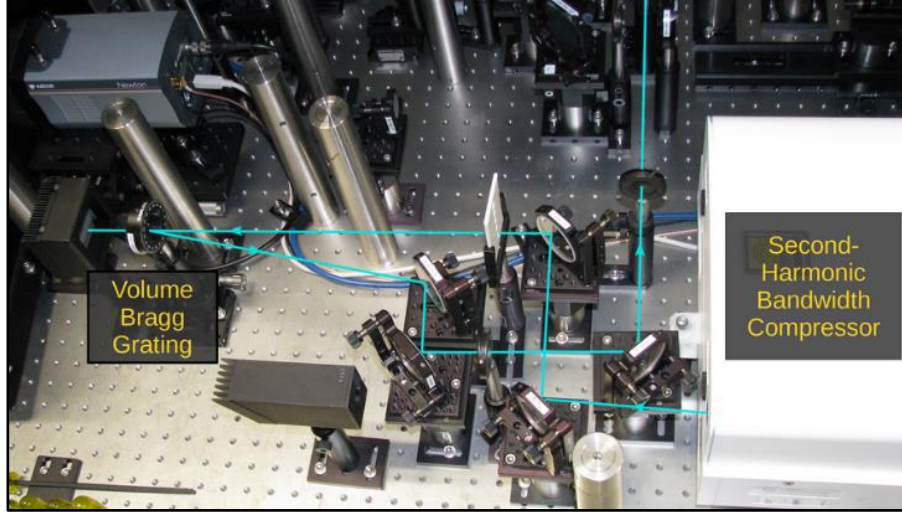


Fig. 5. Annotated photograph showing the arrangement of the volume Bragg grating near the exit of the SHBC to remove spectral wings from the 400-nm probe pulse.

The use of the volume Bragg grating removed the spectral wings from the 400-nm probe beam. Probe spectra recorded with and without the volume Bragg grating are shown in Fig. 6. In addition to removing the spectral wings, the volume Bragg grating also narrowed the probe beam spectral width, as shown by the full-width half-maximum (FWHM) values listed in Fig. 6, which enhanced the spectral resolution of the CARS instrument.

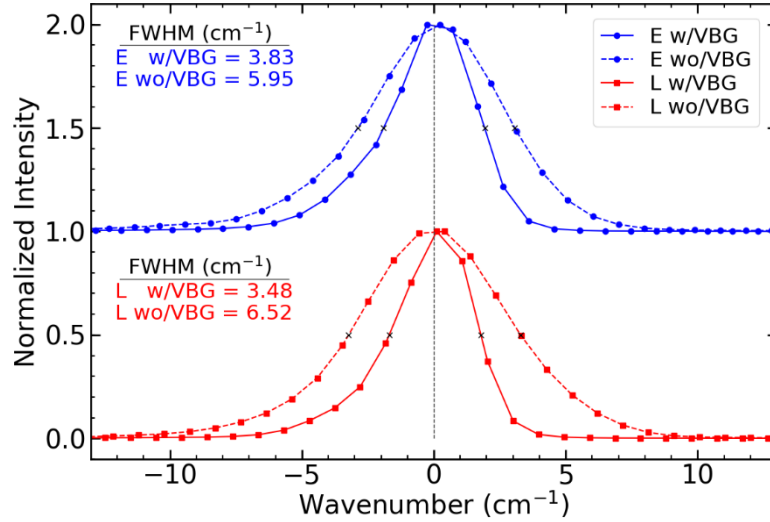


Fig. 6. Probe pulse spectra recorded with (solid lines) and without (dashed lines) the volume Bragg grating. The upper blue lines labeled ‘E’ are the early probe pulse, and the lower red lines labeled ‘L’ are the late probe pulse. The measured full width at half-maximum (FWHM) values are shown in the figure.

The CARS signals generated from the interaction of the early and late probe pulses were spatially separate. A polarization scheme was used to isolate the CARS signals from the intense probe radiation [21], and both CARS signals were directed into a single spectrometer. The two CARS signals were stacked vertically at the entrance to the spectrometer and a single camera (Newton EMCCD, Andor), was used to record both signals at the laser repetition rate of 1 kHz. To achieve this, the camera was run in crop-mode, with the active area reduced to 350 pixels wide by 80 pixels tall; 40 pixels were binned vertically for each CARS signal. The vertical shift speed of the camera read out

was 4.88 μs , and the pixel readout rate was 3 Mhz. The electron multiplying gain function of the camera was not used.

Using a pump energy of 84 μJ , and the volume-Bragg-grating filtered probe pulse, CARS spectra were recorded in the cryostat and fit for temperature and pressure. Single-laser-shot CARS spectra recorded in the cryostat at $T = 104\text{ K}$, $p = 5\text{ Torr}$, with early and late probe delays of 5 and 5000 ps, respectively, are shown along with best-fit theoretical spectra in Fig. 7. Histograms of best-fit temperatures and pressures from 100 such single-shot CARS spectra are shown in Fig. 8.

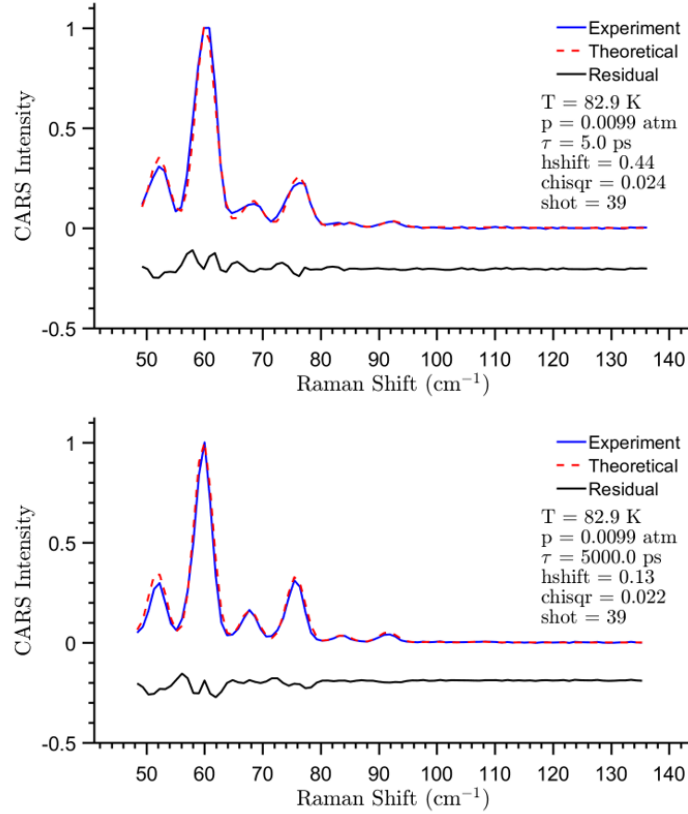


Fig. 7. Single-laser-shot CARS spectra recorded in the cryostat at $T = 104\text{ K}$, and $p = 5\text{ Torr}$ are shown along with best-fit theoretical spectra, the residual of the fit, and fitting parameters including the measured temperature and pressure.

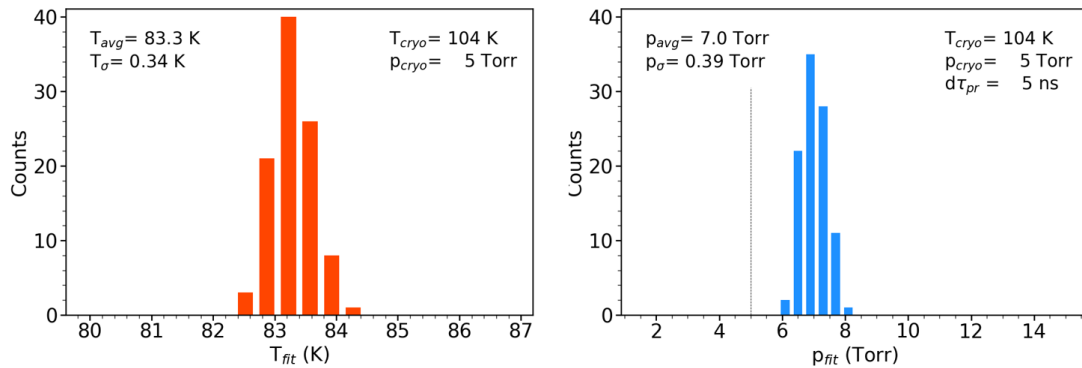


Fig. 8. Histograms of best-fit temperature and pressure from 100 single-laser-shot CARS spectra recorded in the cryostat at $T = 104\text{ K}$, and $p = 5\text{ Torr}$.

The average best-fit temperature for these cryostat conditions was 83.3 K which is significantly lower than the thermocouple-based measurement of 104 K. We suspect that this discrepancy is at least partially a result of heat conduction to the thermocouple from the cryostat housing where the thermocouple was mounted. The cryostat wall temperature was close to ambient temperature. The CARS measurement was performed in close proximity to a liquid-nitrogen-cooled surface, and it is not surprising that the gas temperature would be very similar to that of the liquid nitrogen. The average best-fit pressure was 7.0 Torr which is significantly higher than the convection-gauge measurement of 5 Torr. This discrepancy could be the result of inaccuracies in the Raman J-dependent linewidths at these temperatures. These discrepancies are the subject of continued research.

IV. Conclusion

A fs CARS instrument for temperature and pressure measurements in Sandia National Laboratories' hypersonic wind tunnel is being developed and the current measurement capability has been presented. A broadband fs laser pulse excited many rotational Raman transitions in the gas flow, and the Raman coherence is probed by an early and a late probe pulse. It was found that excessive amounts of pump energy lead to flow perturbations and spuriously elevated CARS temperature data recorded in the hypersonic wind tunnel. Limits on the amount of pump energy were determined using a low-pressure cryostat to simulate wind tunnel conditions. The use of a second-harmonic bandwidth compressor was demonstrated for two-channel CARS spectra acquisition at 1 kHz using a single detection system. A volume Bragg grating was used to spectrally filter the SHBC pulse to remove spectral wings and narrow the ps probe pulse. Simultaneous temperature and pressure measurements were performed in a cryostat at conditions similar to those found in the hypersonic wind tunnel.

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References

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- [1] Casper, Katya, Steven Beresh, John Henfling, Russell Spillers, Brian Pruett, and Steven Schneider. "Hypersonic wind-tunnel measurements of boundary-layer pressure fluctuations." In 39th AIAA fluid dynamics conference, p. 4054. 2009.
 - [2] Casper, Katya M., Steven J. Beresh, and Steven P. Schneider. "Pressure fluctuations beneath instability wavepackets and turbulent spots in a hypersonic boundary layer." *Journal of Fluid Mechanics* 756 (2014): 1058-1091.
 - [3] Lynch, Kyle P., Elizabeth Jones, and Justin L. Wagner. "Simultaneous PSP and surface deformation measurements for fluid-structure interactions in a shock tube." In 2018 Fluid Dynamics Conference, p. 3870. 2018.
 - [4] Casper, Katya M., Justin L. Wagner, Steven J. Beresh, Russell W. Spillers, and John F. Henfling. "Fluid-Structure Interactions on a Tunable Store in Complex Cavity Flow." *Journal of Aircraft* 56, no. 4 (2019): 1501-1512.
 - [5] Currao, Gaetano MD, Andrew J. Neely, Christopher M. Kennell, Sudhir L. Gai, and David R. Buttsworth. "Hypersonic fluid-structure interaction on a cantilevered plate with shock impingement." *AIAA Journal* 57, no. 11 (2019): 4819-4834.
 - [6] Daub, Dennis, Burkard Esser, and Ali Gülhan. "Experiments on High-Temperature Hypersonic Fluid-Structure Interaction with Plastic Deformation." *AIAA Journal* 58, no. 4 (2020): 1423-1431.

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- [7] Sullivan, Bryson T., Daniel J. Bodony, Thomas Whalen, and Stuart Laurence. "Direct Simulation of Fluid–Structure Interaction in a Hypersonic Compression-Ramp Flow." *AIAA journal* 58, no. 11 (2020): 4848-4865.
 - [8] Pandey, Anshuman, and Katya M. Casper. "Hypersonic Fluid-Structure Interaction on the Control Surface of a Slender Cone." In *AIAA SciTech 2021 Forum*, p. 0909. 2021.
 - [9] Parziale, N. J., J. E. Shepherd, and H. G. Hornung. "Differential interferometric measurement of instability in a hypervelocity boundary layer." *AIAA Journal* 51, no. 3 (2013): 750-754.
 - [10] Grisch, F., P. Bouchardy, M. Pealat, B. Chanetz, T. Pot, and M. C. Coet. "Rotational temperature and density measurements in a hypersonic flow by dual-line CARS." *Applied Physics B* 56, no. 1 (1993): 14-20.
 - [11] Foglesong, Robert E., Stephen M. Green, Robert P. Lucht, and J. Craig Dutton. "Dual-pump coherent anti-Stokes Raman scattering for simultaneous pressure/temperature measurement." *AIAA journal* 36, no. 2 (1998): 234-240.
 - [12] Woodmansee, Mark A., Robert P. Lucht, and J. Craig Dutton. "Development of high-resolution N₂ coherent anti-Stokes Raman scattering for measuring pressure, temperature, and density in high-speed gas flows." *Applied Optics* 39, no. 33 (2000): 6243-6256.
 - [13] Kearney, Sean P., and Paul M. Danehy. "Pressure measurements using hybrid femtosecond/picosecond rotational coherent anti-Stokes Raman scattering." *Optics letters* 40, no. 17 (2015): 4082-4085.
 - [14] Escofet-Martin, David, Anthony O. Ojo, Joshua Collins, Nils Torge Mecker, Mark Linne, and Brian Peterson. "Dual-probe 1D hybrid fs/ps rotational CARS for simultaneous single-shot temperature, pressure, and O₂/N₂ measurements." *Optics Letters* 45, no. 17 (2020): 4758-4761.
 - [15] Kearney, Sean P., Daniel R. Richardson, Jonathan E. Retter, Chloe E. Dedic, and Paul M. Danehy. "Simultaneous Temperature/Pressure Monitoring in Compressible Flows using Hybrid fs/ps Pure-Rotational CARS." In *AIAA Scitech 2020 Forum*, p. 0770. 2020.
 - [16] Naik, Sameer V., Waruna D. Kulatilaka, Krishna K. Venkatesan, and Robert P. Lucht. "Pressure, temperature and velocity measurements in underexpanded free jets using laser-induced fluorescence imaging." *AIAA journal* 47, no. 4 (2009): 839-849.
 - [17] Boguszko, Martin, and Gregory S. Elliott. "On the use of filtered Rayleigh scattering for measurements in compressible flows and thermal fields." *Experiments in Fluids* 38, no. 1 (2005): 33-49.
 - [18] Beresh, Steven J., Katya M. Casper, Justin L. Wagner, John Henfling, Russell Spillers, and Brian O. Pruett. "Modernization of Sandia's Hypersonic Wind Tunnel." In *53rd AIAA Aerospace Sciences Meeting*, p. 1338. 2015.
 - [19] Dogariu, Arthur, Laura E. Dogariu, Michael S. Smith, John Lafferty, and Richard B. Miles. "Single shot temperature measurements using coherent anti-Stokes Raman scattering in Mach 14 flow at the Hypervelocity AEDC Tunnel 9." In *AIAA Scitech 2019 Forum*, p. 1089. 2019.
 - [20] Dogariu, Arthur, Laura E. Dogariu, Michael S. Smith, Brianne McManamen, John F. Lafferty, and Richard B. Miles. "Velocity and Temperature Measurements in Mach 18 Nitrogen Flow at Tunnel 9." In *AIAA Scitech 2021 Forum*, p. 0020. 2021.
 - [21] Bohlin, Alexis, Brian D. Patterson, and Christopher J. Kliwer. "Communication: Simplified two-beam rotational CARS signal generation demonstrated in 1D." *The Journal of chemical physics* 138, no. 8 (2013): 081102.
 - [22] Kearney, Sean P., and Daniel J. Scoglietti. "Hybrid femtosecond/picosecond rotational coherent anti-Stokes Raman scattering at flame temperatures using a second-harmonic bandwidth-compressed probe." *Optics Letters* 38, no. 6 (2013): 833-835.
 - [23] Kearney, Sean P. "Hybrid fs/ps rotational CARS temperature and oxygen measurements in the product gases of canonical flat flames." *Combustion and Flame* 162, no. 5 (2015): 1748-1758.
 - [24] Courtney, Trevor L., Nils Torge Mecker, Brian D. Patterson, Mark Linne, and Christopher J. Kliwer. "Hybrid femtosecond/picosecond pure rotational anti-Stokes Raman spectroscopy of nitrogen at high pressures (1–70 atm) and temperatures (300–1000 K)." *Applied Physics Letters* 114, no. 10 (2019): 101107.
 - [25] Castellanos, Leonardo, Francesco Mazza, Dmitrii Kliukin, and Alexis Bohlin. "Pure-rotational 1D-CARS spatiotemporal thermometry with a single regenerative amplifier system." *Optics Letters* 45, no. 17 (2020): 4662-4665.
 - [26] Zhao, Huijie, Ziyang Tian, Tao Wu, Yan Li, and Haoyun Wei. "Dynamic and sensitive hybrid fs/ps vibrational CARS thermometry using a quasi-common-path second-harmonic bandwidth-compressed probe." *Applied Physics Letters* 118, no. 7 (2021): 071107.