High-resolution broadband N₂ coherent anti-Stokes Raman spectroscopy: comparison of measurements for conventional and modeless broadband dye lasers

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We have performed high-resolution N₂ coherent anti-Stokes Raman spectroscopy (CARS) measurements using a modeless dye laser (MDL) as the Stokes beam source to determine the effects of a reduction in mode noise on the accuracy and precision of the method. These results are compared with previous research that employed a conventional broadband dye laser (CBDL) as the Stokes beam source. A new spectral-fitting procedure was developed to avoid starting-point bias in the least-squares fitting results, which possibly had altered the previous measurements. Single-shot measurements of pressure were performed in a static-pressure vessel over the range of 0.1–4.0 atm to examine the pressure sensitivity of the technique. The precision of these measurements is a measure of the baseline noise level of the system, which sets the detection limit for flow-field pressure fluctuations. Centerline measurements of pressure and temperature in an underexpanded jet (\(M_j = 1.85\)) were also used to determine the performance of the technique in a compressible flow field. Our study represents the first known application, to our knowledge, of a MDL CARS system in a low-temperature, low-pressure supersonic environment. Improvements in accuracy for mean single-shot measurements and increased precision were found for pressure vessel conditions above 1.0 atm. For subatmospheric pressure vessel conditions (0.1–1.0 atm) and the underexpanded jet measurements, there was a decrease in accuracy and precision compared with the CBDL results. A comparison with the CBDL study is included, along with a discussion of the MDL system behavior. © 2003 Optical Society of America

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1. Introduction

As the demand for improved performance of high-speed missiles, projectiles, and aircraft increases, so does the requirement for extensive knowledge of the thermodynamic and kinematic properties of the associated compressible flow fields. For the projectile and missile designer, the base drag¹ and infrared plume signature² are important design criteria that are dependent on near-wake properties. Similarly, in aircraft design, structural fatigue due to acoustic fluctuations presents an important issue.³ In addition, other important practical applications, such as supersonic combustion, gas dynamic lasers, and chemical vapor deposition, depend on an in-depth understanding of compressible flows.⁴ Because of compressibility effects in these flows, intrusive probe-based measurements are limited in their accuracy by interference effects. Therefore advances in nonintrusive, laser-based diagnostics are required to assist in the design and implementation of the applications listed above.

In this paper we present the advancement and application of a broadband coherent anti-Stokes Raman spectroscopy (CARS) technique previously used to obtain pressure and temperature measurements in a compressible flow. As we discuss in detail, the conventional broadband dye laser (CBDL) utilized in previous research was replaced with a modeless dye laser (MDL) to reduce the baseline noise level seen in

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associated pressure vessel measurements. This noise level, which is evident in a quiescent environment, sets the detection limit for flow-field pressure fluctuation measurements. The system performance is then examined in a low-temperature, low-pressure underexpanded jet, which is a typical and important supersonic flow.

A brief discussion of the current CARS technique is presented here; a more thorough review of the theory can be found in the extensive reviews of CARS that exist in the literature. For the measurements discussed in this paper, the first vibrational Raman transition \((v = 0 \rightarrow 1)\) of diatomic nitrogen is probed through a third-order, nonlinear wave-mixing process. Two degenerate pump beams \((v_p)\) and a Stokes beam \((v_S)\) are focused and overlapped in the probe volume, where the CARS signal \(v_{\text{CARS}} = 2v_p - v_S\) is generated. The source for the Stokes beam is broadband, allowing for multiple transitions to be probed simultaneously and for instantaneous, or single-shot, measurements of pressure and temperature to be acquired. Pressure and temperature are determined from the spectral shape of the signal. For this study, software known as CARSSPT, developed at Sandia National Laboratories, is employed to perform the least-squares fitting process. To generate our theoretical spectra, the collisionally broadened linewidth relations of Rahn and Palmer were employed.

Because the process relies on molecules already present in the flow, there is no need for tracer molecules or particles, such as required by fluorescence-based techniques or laser Doppler velocimetry. Thus this laser-based method is nonintrusive and nonperturbing. The coherent nature of the signal provides a high signal-to-noise ratio compared with incoherent light-collection techniques such as spontaneous Raman scattering. With use of the folded-boxcars phase-matching geometry, the signal is spatially separated from the input beams. The geometry used here provides an ellipsoidal probe volume approximately 1300 \(\mu\)m long by 40 \(\mu\)m in diameter. When the probe volume is located at the centerline of the underexpanded jet (most severe condition), the maximum differences in pressure and temperature from the center of the ellipsoid representing the probe volume to the vertex are 2% and 5%, respectively. Thus the geometry provides sufficient spatial resolution to probe the underexpanded jet to within the error limits of the system. The CARS process implemented here employs visible wavelengths, thus alleviating the issues involved in use of ultraviolet wavelengths required of other techniques such as laser-induced fluorescence. Because of the relatively high signal-to-noise ratio, the CARS process also provides the ability to probe areas of low molecular number densities.

### 2. Previous Research

The development of CARS for measurements in high-speed flows has been ongoing at the University of Illinois for several years. The research started with dual-pump CARS\(^{12}\) measurements of pressure and temperature. In the experiments of Foglesong et al.,\(^{13}\) the vibrational and pure rotational transitions of diatomic nitrogen were probed simultaneously in a stagnant gas cell and along the centerline of an underexpanded jet. The technique provided accurate time-averaged pressure measurements above 1 atm, as well as accurate mean and instantaneous temperature results throughout the range of 100–300 K. Unfortunately, the method performed poorly for time-averaged pressure measurements below 1 atm and for instantaneous pressure measurements below 3 atm. For instantaneous pressure measurements, the standard deviation was reported at approximately 10% of the mean for 3 atm, dropping to 4% at 18 atm. As this baseline noise level is of the order of the fluctuations expected in a typical compressible flow field, the pressure fluctuation measurements obtained with the technique would not be representative of actual flow variations.

In light of these results, modifications to the dual-pump CARS technique were made to increase pressure sensitivity at lower pressure and to investigate the ability of the method to obtain accurate, instantaneous, single-shot measurements. Instead of probing both pure rotational and vibrational resonances of diatomic nitrogen, the modified technique focused on vibrational resonances. Whereas in the previous study the CARS signal spanned a range between 40 and 100 \(\text{cm}^{-1}\), the modified CARS measurements spanned only 10 \(\text{cm}^{-1}\). Using a relay lens system to magnify the CARS signal spectrally, with this new high-resolution technique we were able to resolve the CARS signal to within 0.10 \(\text{cm}^{-1}\), allowing pressure and temperature sensitivity of the diatomic nitrogen Q branch \((v = 0 \rightarrow 1)\) to be investigated. As before, the method was examined in a stagnant gas cell and throughout the flow field of an underexpanded jet. The results from the gas cell revealed that only below 2 atm was the method capable of making accurate time-averaged and ensemble-averaged single-shot measurements of mean pressure, therefore displaying the opposite pressure sensitivity as compared with the previous dual-pump method. The high-resolution technique also provided accurate mean pressure and temperature measurements in the underexpanded jet flow field. Even at the low molecular number densities encountered in the jet flow field, the method displayed the ability to generate high signal-to-noise on a single-shot basis such that temperature and pressure could be deduced.

One goal of this modified technique was to provide the ability to measure fluctuating thermodynamic properties in compressible shear flows.\(^{14}\) Although the technique could provide single-shot measurements with accurate mean values, the ensemble-averaged fluctuations were dominated by inherent system noise. From the gas cell experiment, the baseline noise level over the 0.1–5.0-atm range examined was approximately 12% of the mean pressure value on average. These artificial fluctuations (precision uncertainty), which dominated the results,
were attributed to mode-noise contributions from the CBDL that was used as the source for the Stokes beam.\textsuperscript{15}

In the current study we examine modifications to the high-resolution CARS method to extend its capabilities to measure mean and fluctuating thermodynamic properties. To reduce the effects of mode noise, a MDL, based on the design of Ewart,\textsuperscript{16} was implemented as the new source for the Stokes beam. The diffraction grating was removed from the MDL in this setup, thus avoiding the spatial variation issues encountered by Snelling \textit{et al.}\textsuperscript{17} and subsequently increasing the conversion efficiency of the MDL. The capabilities of the method were benchmarked in two well-defined experiments. First, the pressure sensitivity and the system noise level were assessed in a gas cell. Then we obtained measurements along the centerline of an underexpanded jet flow field. The results of these investigations were used to determine the accuracy and precision of the new CARS method as compared with the previous CBDL study.

3. Mode Noise

The major drawback to one using a CBDL as a Stokes beam source is the oscillator cavity. Use of the cavity requires that a given frequency oscillate only if it forms a standing wave within the cavity. These frequencies, or axial modes, will modulate the spectral profile of the CBDL, which in turn alters the spectral profile of the CARS signal and the measurements derived from it. An in-depth discussion of mode noise and its effects on the CARS method can be found in a study by Greenhalgh and Whittley.\textsuperscript{18}

Because the source for the mode noise results from the oscillator cavity, two new dye laser designs that do not rely on a cavity have been developed. These two designs have been used almost exclusively at combustion temperature levels, with no thorough investigations being performed at low temperature to our knowledge. The first design was developed by Ewart,\textsuperscript{16} for which stimulated emission is amplified in four stages without oscillation. Since its inception, this MDL has been utilized in a few CARS investigations, two of which are reviewed here. Snowdon \textit{et al.}\textsuperscript{19} report a reduction in the standard deviation of noise of single-shot CARS spectra from $\sim$13\% for a CBDL to $\sim$5\% for a MDL, resulting in improved precision of single-shot temperature measurements. Use of the MDL provided a reduction from $\sim$5\% to $\sim$1.25\% in standard deviation of temperature at a mean value of 1200 K. In a second investigation a systematic evaluation of different styles of dye lasers was performed.\textsuperscript{17} The comparisons revealed a reduction from $\sim$22\% to $\sim$6\% in standard deviation of CARS signal noise when a CBDL was switched to a MDL. The investigation also displayed increased precision of single-shot temperature measurements, as standard deviations dropped from $\sim$3.5\% to $\sim$1.5\% at a mean temperature of 1600 K.

The second MDL design, introduced by Hahn \textit{et al.},\textsuperscript{20} makes use of Bethune dye cells.\textsuperscript{21} The Hahn \textit{et al.} report\textsuperscript{20} includes an evaluation of the capabilities of the MDL design. Although percent standard deviation of CARS signal noise was not reported, reduction in percent standard deviation of temperature measurements was. Use of this MDL reduced temperature fluctuations from $\sim$5\% to $\sim$2\% at 1200 K. This MDL was also employed in a separate investigation. Lucht \textit{et al.}\textsuperscript{22} utilized this MDL design in the measurement of temperature and CO\textsubscript{2} concentration. No comparison was given to experiments with a CBDL, but the percent standard deviation of temperature was reported at $\sim$2\% for a mean temperature of 2000 K. These four MDL studies solidify the benefits of a MDL over a CBDL, as the findings of significant reductions in noise levels in CARS signal intensity and resulting temperature measurements are consistent.

Although these percentage standard deviations are low enough to resolve a typical probability density function in a turbulent combustion environment,\textsuperscript{5} this precision may not be enough for meaningful single-shot measurements in a turbulent, supersonic flow field. It should be noted that the lowest percentage standard deviation reported above corresponds to an absolute standard deviation of 15 K, which is a significant level for a typical supersonic flow for which mean temperature levels of the order of 100 K occur. Because there is no indication as to whether the above success in noise reduction will scale in an absolute or percentage sense for low-temperature flows, an estimate for how a MDL CARS method will perform at supersonic flow conditions cannot be made. In addition, there is no clear method of correlating the decrease in standard deviation of temperature to one for pressure, which is investigated in this study. Therefore the following results represent to our knowledge the first investigation of the effects of MDL on the precision of CARS pressure and temperature measurements in a high-speed, nonreacting environment.

4. Experimental Facilities and Equipment

A top view of the CARS system is shown in Fig. 1. The Q-switched, injection-seeded Nd:YAG laser (Continuum Powerlite 8010) has a maximum pulse energy of approximately 900 mJ at a wavelength of 532 nm for a pulse duration of $\sim$6 ns. The beam is directed through a quarter-wave plate and a Glan polarizer; these two optics comprise the power attenuation control for the balance of the CARS system. A beam splitter separates 20\% of the beam intensity for the pump beams in the CARS process ($v_p$). The pump beam is directed through a delay line so that it arrives at the probe volume coincident in time with the Stokes beam. Shortly before the beam is directed to the probe volume, it is split equally by a 50\% beam splitter to form the two necessary pump beams.

The remaining 80\% of the initial 532-nm beam is used to pump the MDL. As stated above, the MDL is derived from the Ewart\textsuperscript{16} design, modified to include an additional amplification stage. Rhodamine 640 perchlorate dissolved in methanol provides the gain medium. The concentration in the initial dye
The cell was 22 mg in 400 ml and in the remaining dye cells was 16 mg in 400 ml. The MDL emits coherent light in the range of 607 nm at approximately 25 mJ/pulse and is the source for the Stokes beam in the CARS process ($v_\text{S}$). The Stokes beam is then directed into the probe volume. The last turn made by each of the three input beams is performed with a prism fixed to a high-precision mirror mount (Newport 610 Series) to allow for fine-tuning of the probe volume alignment. The arrival times of the three input beams are checked at this point in the system with a fast photodiode and oscilloscope to ensure that all beams reach the probe volume within 1 ns of each other. The translation system is based on an earlier, successful design, and has a positional repeatability of less than 5 μm in each direction. This allows for probe volume movement without beam realignment at every measurement location.

The CARS signal is then directed to the 1.25-m spectrometer (SPEX 1250M); see Fig. 1. A 100-mm lens focuses the CARS signal onto the entrance slit, which is set to a width of 150 μm. The spectrometer design is based on a single-pass Czerny–Turner configuration with a 3600-groove/mm holographic grating. This configuration provides a theoretical resolution of $\Delta \nu \approx 0.09$ cm$^{-1}$. At the exit of the spectrometer, a relay lens pair disperses the signal onto the unintensified CCD (Roper Scientific NTE/2500PB). The CCD is a 2500 × 600 pixel, 16-bit array that provides on average one count for every 3.5 incident photons. The relay lens pair consists of 28- and 210-mm focal-length Nikon camera lenses focused at infinity, resulting in a magnification factor of 7.5. The image of the CARS signal is captured and binned on the CCD chip to provide the CARS spectra that will be analyzed.

An optically accessible gas chamber was modified for use in this study. Pressure is monitored independently of the CARS measurements with digital pressure transducers: a 1000-Torr absolute transducer (MKS 722A13TCE2FK) for subatmospheric studies and a 5000-Torr absolute transducer (MKS 722A53TCE2FK) for above-atmospheric studies. Both transducers are accurate to 0.5% of the pressure reading.

The underexpanded jet facility is similar in design to previous investigations, except here the nozzle exit diameter is doubled. Dry, high-pressure air is supplied to the horizontally mounted inlet pipe at approximately 9 atm. The stagnation pressure and temperature are monitored with the 5000-Torr transducer described above and a hermetically sealed thermistor (Omega ON-920-4007), respectively. The nozzle exit is 10 mm in diameter, and the nozzle diameter contracts linearly from the inner supply pipe diameter of 19.3 mm to the exit. The area reduction, which occurs over 101.6 mm, results in an angle of contraction of 2.61°. The jet can be operated over a fully expanded jet Mach number $M_j$ range up to ~2. In the present study, the jet was operated at a nozzle pressure ratio ($P_0/P_\text{amb}$) of 6.17, which corresponds to $M_j = 1.85$.

5. Spectral-Fitting Procedure

When analyzing data for the pressure vessel, the final answer is known a priori from the transducer measurements. However, in the flow field investigations, the pressure and temperature measured are not known before the spectra are fit. As CARSFIT requires an initial value for the floated variables, an estimate of these conditions must be made. If an individual spectrum is of poor quality or if the specified initial values of the fit are too far from the true conditions, CARSFIT may return the initial values as an approximation.
indication that a fit could not be successfully performed. Therefore a procedure was developed to determine if the returned values were an accurate prediction of pressure and temperature or if the spectrum should be discarded. This helps to identify the effects of starting-point bias. The procedure was tested on sample pressure vessel data to determine its effectiveness.

We begin the process by initially discarding any spectra that exhibited a saturated CCD pixel or contained a low integrated signal intensity. The remaining spectra are fit starting from three unique points, based on known or expected values. If two of the three fitting results are within a set percentage of each other, the spectrum with the lowest chi-squared, goodness-of-fit value is retained, regardless of the prediction of temperature or pressure. However, if all three fitting results are separated by more than the set percentage, the spectrum is discarded. For the pressure vessel, for which only pressure is allowed to float, each spectrum was started at 70%, 100%, and 130% of the transducer-measured pressure and was retained if two of the results were within 10% of each other. In the case of a flow field for which both pressure and temperature are allowed to float, both pressure and temperature must simultaneously meet this criterion for the spectrum to be retained.

Using 70% and 130% starting points for comparison considerably relaxes the required accuracy when we estimate the thermodynamic conditions at the probe volume for use as initial values in the spectral-fitting procedure. However, this advantage in turn brought about the concern that CARSFIT may not be capable of starting this far from the true condition and still be able to return an accurate prediction. Hence an experiment was performed on a small set of time-averaged pressure vessel spectra for which 70% and 130% starting points were used, as well as 90% and 110% starting points for comparison. As is evident in Fig. 2, the mean pressure levels resulting from both sets of starting values are in excellent agreement. These results verify the utility and accuracy of the newly developed spectral-fitting procedure and display the reliability of the pressure and temperature measurements obtained with this data-reduction method.

6. Pressure Vessel Results

The current results represent 500 single-shot measurements acquired from the gas cell at 25 pressure levels in the range from 0.1 to 4.0 atm. Figure 3 displays single-shot CARS spectra obtained in the pressure vessel at 0.1, 1.0, and 3.0 atm and 292 K along with the theoretical results from CARSFIT. Each spectrum is labeled with the transducer pressure, $P_{\text{TRAN}}$, and the predicted CARSFIT pressure, $P_{\text{CARS}}$. As CARSFIT compares the square root of intensity in the least-squares fitting process, all subsequent results are plotted as such.

All spectra in Fig. 3 display excellent agreement between data and theory. Not only are the peak

Fig. 2. Comparison of the mean spectral-fitting procedure results with different starting points.

Fig. 3. Comparison of single-shot experimental CARS and theoretical CARSFIT spectra from the pressure vessel for (a) 0.1 atm, (b) 1.0 atm, and (c) 3.0 atm.
intensities of each transition well represented by the theory, but the linewidth and line shape are as well. It is important to note that all theoretical spectra presented here were generated at a resolution of 0.10 cm\(^{-1}\), verifying that the experimental spectra were detected at high resolution. The linewidths of the transitions broaden and interfere as pressure increases, and the resulting spectrum is determined by the competing effects of collisional broadening and collisional narrowing.\(^{25}\) It is these features that give the technique its pressure sensitivity. This sensitivity is evident, as the results obtained in Figs. 3(b) and 3(c) agree with those from the transducer to within 2%.

The low signal level that can be seen in Fig. 3(a) for 0.1 atm is a cause for concern. Because the experimental setup closely resembles the previous setup\(^ {11}\) in all ways aside from the MDL that is used in place of a CBDL, the MDL is considered the most probable cause for this low signal level. Although the total average power out of the MDL is 30% lower than previous experiments with the CBDL,\(^ {11}\) this does not completely account for the drop in signal level. Considering all the differences between the two experimental setups, there is still an order-of-magnitude less signal generated in the current system. The low signal level is most likely attributed to the decrease in spatial beam quality and transverse electromagnetic-mode (TEM) structure provided by the MDL as compared with the previous CBDL, which employed a fixed-length oscillator cavity. This degradation in beam quality lowers the efficiency at which the CARS signal is generated in the nonlinear wave-mixing process. This will increase the effect of shot noise on the CARS spectra generated with the MDL.

A comparison of the mean CARS-predicted pressure and transducer pressure over the entire range of the pressure vessel study is shown in Fig. 4(a), with Fig. 4(b) highlighting the subatmospheric portion of the results. Included in Fig. 4 are the results from the previous study,\(^ {11}\) which is used as a comparison for the current technique. Here (and in the plots for Fig. 9) the uncertainty bars denote the standard deviation of the pressure levels obtained from the ensemble of spectra at each pressure level. Above 1.0 atm, the current results compare well with the transducer values with a 40% average improvement in accuracy over the previous study. As can be seen in Fig. 4(b), the accuracy of the technique decreases for pressure levels below 1.0 atm, with a majority of CARS values that are below the transducer values. Because the transducer pressure for the subatmospheric region was monitored with different transducers and the results were similar, it is unlikely that this loss of accuracy is due to a zero offset bias of the transducer. In addition, this difference is not attributable to an inaccurate spectral instrument function, as the one employed in the convolution of the theoretical spectra was varied to provide the best comparison between experimental and theoretical spectra and the most accurate mean results. This decrease in accuracy is therefore most likely the indirect result of the decrease in signal level at these lower molecular number density levels. This discrepancy is unfortunate, as the results for pressure levels above 1.0 atm indicate that the inclusion of the MDL in the system improves the accuracy of the technique.

Figure 5 displays the precision of the technique when we plot the standard deviations of the single-shot measurements for the corresponding pressure level. In comparison with the previous study,\(^ {11}\) the increased accuracy of the technique above 1.0 atm is complemented by a relatively small but discernible increase in precision as well. The increase in standard deviation below 1.0 atm compared with the previous results is not unexpected, as it is believed that the previous measurements were affected by the starting-point bias discussed above. In addition, the decrease in CARS signal strength degrades current system performance at low pressure. The number of spectra retained from the ensemble of 500 single-shot spectra at each pressure level is presented in Fig. 6.
As the technique loses pressure sensitivity with increasing pressure, there is a decrease in the number of spectra retained above 2.0 atm. Still, nearly all the single-shot spectra obtained were successfully fit from at least two different starting points with best-fit pressure values falling within the prescribed tolerance. Not only were these spectra fit successfully, but in addition, each data set provided accurate mean pressure values, as can be seen in Fig. 4. This result confirms the ability of the technique to provide accurate predictions of pressure when the actual conditions at the probe volume are not known.

7. Underexpanded Jet Results

The pertinent flow features of an underexpanded jet are shown in Fig. 7. For an underexpanded jet, the exit pressure of the nozzle is greater than that of the ambient surroundings. This pressure mismatch causes a Prandtl–Meyer expansion fan to form at the lip of the nozzle. These expansion waves propagate across the flow field and reflect from the constant-pressure jet boundary as compression waves, which in turn coalesce to form the intercepting shock (or barrel shock) that is attached to the nozzle lip. The inviscid core flow is accelerated to supersonic speeds by the expansion fan, traveling downstream to a point where the intercepting shock would cross the centerline. Here, the flow is recompressed. If the exit pressure is great enough (a nozzle pressure ratio greater than or equal to 3), the simple crossing (i.e., regular reflection) of the intercepting shock along the centerline will no longer provide sufficient pressure rise, and a minimally curved normal shock, or Mach disk, forms. Instead of crossing (reflecting) at the centerline, the intercepting shock now attaches to the edge of the Mach disk, where the reflecting shock forms and propagates downstream to the constant-pressure boundary. The triple point is located at the intersection of the intercepting shock, reflecting shock, and Mach disk and is the starting point for the slip line or inner shear layer. The inner shear layer is concentric with the outer shear layer, which resides along the constant-pressure boundary. As the reflecting shock reflects from the outer shear layer, a new series of expansion waves forms. A shock train forms, similar in geometry to the first shock cell, and continues downstream until the pressure mismatch is dissipated by viscosity. As in the subsonic case, the outer and inner shear layers will grow until they overrun the jet core region, and the jet becomes fully turbulent.

In this investigation, 250 single-shot spectra were obtained at nine locations along the centerline from the jet exit to just beyond the Mach disk. Figure 8 displays single-shot spectra at centerline positions corresponding to the jet exit, halfway to the Mach disk, just upstream of, and just downstream of the Mach disk ($z/d_j = 0.019$, 0.870, 1.508, and 1.579,
respectively) along with the fitted theoretical spectra. Included in the plots are the CARSFIT ($P_{\text{CARS}}$ and $T_{\text{CARS}}$) and computational fluid dynamics (CFD) ($P_{\text{CFD}}$ and $T_{\text{CFD}}$) predictions of pressure and temperature for comparison. The details of the CFD simulation are discussed in the previous study. As with the pressure vessel results, the experimental spectra are well represented by the theoretical spectra, having a coefficient of determination of 0.96 on average. Special attention is drawn to the differences between Figs. 8(c) and 8(d), where the large differences in thermodynamic conditions upstream and downstream of the Mach disk (nominally 0.25 atm and 100 K to 1.25 atm and 250 K, respectively) are readily apparent in the spectra. The increase in temperature results in the diatomic nitrogen population being redistributed from, approximately, the first 20 rotational levels to the first 30, and the linewidths are broadened by a factor of 4 due to the rise in pressure. These spectra illustrate the sensitivity of the technique to these different thermodynamic conditions. Moreover, for all the spectra, there is a close correlation between the CFD calculations and experimental measurements of pressure and temperature, as they agree on average to within 2% and 11%, respectively. In general the CFD calculations and experimental measurements of pressure are in better agreement than is the case for temperature. This result is in contrast to the previous study where temperature predictions were more accurate. The increased discrepancy in temperature predictions that can be seen in Figs. 8(c) and 8(d) can be accounted for when we consider the unsteady position of the Mach disk, which will add to experimental error at these locations, along with the inability of the CFD to accurately capture the normal shock location.

The mean results from the single-shot spectra at all centerline locations examined are compared with previous measurements and the CFD results in Fig. 9. Clearly, there is a substantial improvement in agreement between the CARS measurements and the CFD results for pressure levels above 1.0 atm. This result is expected from the pressure vessel measurements. There is a small
but consistent discrepancy between the CFD and the experimental mean-temperature results upstream of the Mach disk, with the CARS results always being lower. This nearly constant offset of the CARS results is most likely due to the stagnation temperature in the current experiment (290 K) being slightly lower than for the previous study (296 K). The thermodynamic conditions along the centerline will closely follow isentropic expansion predictions for the early stages of jet development. Therefore the temperature measurements between the two experiments should scale with the isentropic $T/T_0$ relation. Comparing the results in this sense reduces the error of the current measurements by 1% on average. Just downstream of the jet exit where the isentropic assumptions are best applied, the scaling reduces the error to less than 1%. Overall, the current time-averaged measurements correlate well with the CFD predictions from the jet exit through the Mach disk.

The standard deviation of the current and previous single-shot measurements, which is representative of the precision of the method, is plotted in Fig. 10. Because there is low turbulence intensity along the centerline of the jet before the Mach disk, the standard deviation results here should represent the baseline detection limit of fluctuations almost exactly. There is a noticeable increase in standard deviation in both pressure and temperature as compared with the previous study. The most plausible explanation for this increase is due to the low signal strength and increased shot noise of the current measurements. Adding temperature as another variable to the least-squares fitting process, as compared with the pressure vessel data, further demonstrates the issues with low CARS signal strength. This is again illustrated in Fig. 11, where the number of spectra retained decreases sharply in the region of low molecular number density just upstream of the Mach disk. The number of fitted spectra it is also shown in this plot, as some of the spectra were discarded before fitting because of saturated pixels or low signal conditions.

Fig. 9. Comparison of mean single-shot centerline results with those of Woodmansee et al. and CFD results for (a) pressure and (b) temperature.

Fig. 10. Comparison of single-shot standard deviation centerline results with those of Woodmansee et al. for (a) pressure and (b) temperature.
8. Conclusions

Reviewing the pressure vessel results, an improvement in accuracy of ensemble-averaged single-shot measurements was shown as compared with previous research for pressure levels above 1.0 atm. This result demonstrates the benefit of one employing the MDL as the Stokes beam source. Decreased performance for subatmospheric pressure levels was attributed mainly to the low CARS signal strength at these conditions. A decrease in standard deviation for the single-shot spectra was evident at above-atmospheric pressure levels, representing increased precision resulting from use of the MDL. Below 1.0 atm, the single-shot standard deviations increased as compared with previous results. Single-shot CARS spectra displayed excellent spectral behavior, allowing for good agreement between theoretical and experimental spectra.

Mean single-shot results from the centerline traverse of an underexpanded jet displayed excellent agreement with CFD results, especially for pressure levels above 1.0 atm. Slight offsets in mean temperature were observed and explained. Experimental spectra from the centerline traverse provided good measurements of pressure, with a minor decrease in agreement in temperature as compared with previous results. Increases in standard deviations for both temperature and pressure occurred, which are most likely indicative of low signal strength. The slight increase in precision for pressure vessel measurements above 1.0 atm confirms that the baseline noise level, or detection limit of flow-field pressure fluctuations, of the technique can be lowered. This could allow for fluctuation measurements to be made with future modifications to the CARS method. The decrease in signal strength at lower pressure levels degraded system measurement precision from that seen in the previous experiment.11

To improve the performance of the current technique, a new source for the Stokes beam should be explored. A promising source would be a MDL based on the design of Hahn et al.20 This laser employs Bethune dye cells21 as opposed to the transversely pumped dye cells in the current MDL, with a single concentrated pass through the oscillator dye cell rather than four distributed passes. As seen in a previous CARS experiment,22 this dye laser provides low standard deviations of temperature measurements in a steady laminar flame. Another possible solution would be to incorporate two single-mode, solid-state tunable sources into the system: one to replace the Stokes source and one to replace a pump beam in a dual-pump CARS technique.12 With the correct choice of wavelengths for these two sources, two transitions of the nitrogen Q-branch could be probed simultaneously. This second option is less attractive as it involves a potential decrease in pressure sensitivity because it does not probe the entire linewidth of both transitions, and it is also more expensive to implement. Nevertheless, use of single-mode sources for the pump and Stokes beams would provide excellent beam quality and TEM structure and would virtually eliminate mode noise.

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