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Dual-Pump Coherent Anti-Stokes Raman Scattering for Simultaneous Pressure/Temperature Measurement

Robert E. Foglesong,* Stephen M. Green,† Robert P. Lucht,‡ and J. Craig Dutton§
University of Illinois at Urbana–Champaign, Urbana, Illinois 61801

A new dual-pump coherent anti-Stokes Raman scattering (CARS) technique for the simultaneous measurement of pressure and temperature is discussed. For these measurements the wavelengths of the two pump lasers and the Stokes laser are selected so that the vibrational and pure rotational spectra of nitrogen are simultaneously acquired. Because collisional narrowing is an important effect for the vibrational spectrum but not for the isolated pure rotational lines, the relative intensities of the vibrational and pure rotational spectra change markedly as the pressure changes. Consequently, pressure as well as temperature can be determined from the spectral shape of the dual-pump CARS spectrum. The pressure measurement capability of the technique was demonstrated in a room-temperature gas cell at pressures ranging from 0.1 to 20 atm. The pressures determined from averaged dual-pump CARS spectra are in good agreement with pressure transducer measurements over this entire pressure range, with an average absolute error of less than 8%. Simultaneous pressure and temperature measurements were also performed along the centerline of an underexpanded jet. Results agreed well with theoretical predictions. Single-laser-shot pressure and temperature measurements are discussed.

Introduction

DEVELOPMENT of nonintrusive techniques for the measurement of pressure, temperature, and density is essential for the further understanding of turbulent supersonic flowfields. Current modeling efforts are limited by the lack of experimental data against which the models can be benchmarked. Coherent anti-Stokes Raman scattering (CARS) is potentially an excellent technique for nonintrusive measurements in supersonic flowfields because of its excellent temporal resolution and good spatial resolution. CARS measurements can be performed on the nitrogen and/or oxygen molecule and, therefore, the technique requires no seeding, which can be very difficult in high-flow rate supersonic wind tunnels. CARS has been most widely used for measuring temperature and species concentration for a variety of flow applications.[1] The measurement of flow velocity using CARS has also been proposed.[2] In this paper we describe a new technique for the simultaneous measurement of pressure and temperature using dual-pump CARS to simultaneously acquire the vibrational and pure rotational spectra of the nitrogen molecule.

Pure rotational CARS was first reported by Barrett,[3] who observed the pure rotational Raman transitions of hydrogen. Beattie et al.[4] then applied the technique to the lower frequency pure rotational transitions of the oxygen and nitrogen molecules in air. The first use of pure rotational CARS of nitrogen as a technique for measurement of low temperatures was performed by Goss et al.[5] Murphy and Chang[6] then applied the technique on a single-shot basis to obtain time-resolved temperature measurements. The spectral shape of the nitrogen Q branch has been widely used for high-temperature measurements in combustion systems. However, even though the collisional narrowing effects of pressure on the spectral shape of the nitrogen Q branch has been studied extensively,[1] to our knowledge this spectral shape has not conversely been used for the measurement of pressure.

Using the dual-pump technique demonstrated by Lucht et al.,[7] the vibrational and pure rotational spectra of nitrogen can be simultaneously acquired. At the pressures studied here, the isolated nitrogen rotational lines are not susceptible to the effects of collisional narrowing. As a result, the relative intensities of the Q branch and rotational lines change considerably as pressure changes. Thus, by simultaneous acquisition of the vibrational and pure rotational spectra of the nitrogen molecule, pressure and temperature can be measured simultaneously on single-laser shots.

Bengtsson et al.[8] simultaneously acquired vibrational and pure rotational spectra using dual-broadband CARS (DBCARS) with a double-folded BOXCARS phase-matching scheme. However, they focused their efforts on the simultaneous measurement of temperature and concentrations of fuel, oxygen, and nitrogen. The pressure sensitivity of the spectra was not explored. Although this technique gives very similar spectral information to the dual-pump technique discussed here, the double-folded BOXCARS phase-matching scheme, which requires an additional mirror arrangement within the spectrometer, seems to be much more complicated experimentally than the dual-pump technique, for which the rotational and vibrational spectra appear in the same spectral region. The primary advantage of the DBCARS technique is that it requires only a pump laser and a single broadband laser. As will be discussed, the dual-pump CARS system utilizes a pump laser, a narrow-band laser, and a broadband laser.

Simultaneous measurement of temperature and density using a dual-line CARS (DLCARS) technique was explored by Péalat and Lefebvre[9] and Grisch et al.[10] In this technique, two narrow-band dye lasers and a single-frequency pump laser are used to probe two rotational lines of the Q branch of nitrogen (note that these are not pure rotational lines). With this technique the rotational temperature is deduced by comparison of the intensities of the two lines. Because only two lines are used for the determination of temperature, shot-to-shot laser intensity fluctuations generally prohibit use of this technique on a single-shot basis. To compensate for this, Péalat and Lefebvre[9] simultaneously used the DLCARS system to acquire a second spectrum from a cell containing a sample of the gas under study to which the system can be referenced. Absolute intensity can also be referenced to this calibration spectrum to obtain number density in the experimental system. Because of the difficulties mentioned in Ref. 9 relating to consistency in beam overlap between the two probe volumes, it seems that the accuracy of this number density measurement technique is limited. The advantage of this dual-line

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*Graduate Research Assistant, Department of Mechanical and Industrial Engineering. Student Member AIAA.

†Graduate Research Assistant, Department of Mechanical and Industrial Engineering; currently Member of the Technical Staff, The Aerospace Corporation, 2350 East El Segundo Boulevard, El Segundo, CA 90245-4691. Member AIAA.

‡Professor of Mechanical and Industrial Engineering. Senior Member AIAA.

§W. Grafton and Lillian B. Wilkins Professor of Mechanical Engineering. Associate Fellow AIAA.

technique is higher signal intensity compared to the broadband techniques discussed earlier, which allows single-shot measurements to be made at the extremely low densities found in hypersonic flowfields, the focus of Péalat and Lefebvre's investigation.[9]

Laser-induced fluorescence (LIF),[11–13] filtered Rayleigh scattering,[14,15] and stimulated Raman spectroscopy[16] have been used for the simultaneous measurement of temperature, pressure, and velocity. LIF measurements of temperature, pressure, and velocity have been performed on nonreacting flows seeded with NO (Ref. 11) or iodine (Ref. 12) and on reacting flows by the fluorescence of OH (Ref. 13). The primary advantage of CARS over LIF techniques is that CARS does not require nonreacting flows to be seeded with a fluorescing trace species. Thus, CARS is an excellent diagnostic technique for supersonic wind tunnels, in which the high flow rates make seeding with NO or iodine largely impractical. Also, in reacting flows, CARS measurements are not restricted to high-temperature regions of the flow where radical species concentrations are high.

For time-resolved, simultaneous measurement of multiple parameters using filtered Rayleigh scattering, the scattering must be imaged through multiple filters with different absorption profiles or observed from multiple angles simultaneously.[14] Aside from being very complex experimentally, simultaneous acquisition of multiple Rayleigh scattering images on a time-resolved basis is limited by the low signal levels typical of single-shot laser diagnostic measurements.

The dual-pump CARS technique that we discuss further has the advantage that the pressure and temperature are calculated from spectral shapes and, thus, the measurements are relatively insensitive to effects such as laser beam absorption or signal attenuation. LIF, Rayleigh scattering, and stimulated Raman measurements of pressure and temperature are complicated significantly by such effects. However, LIF and Rayleigh scattering in their planar forms offer a distinct advantage over the CARS technique that we discuss in that they provide planar rather than pointwise measurements.

We have demonstrated the mean and time-resolved pressure measurement capability of the dual-pump CARS technique by performing measurements in a room-temperature gas cell at pressures ranging from 0.1 to 20 atm. The capability of the technique for the simultaneous measurement of pressure and temperature has been demonstrated in an underexpanded jet.

Principle

Dual-Pump CARS

Simultaneous detection of vibrational and pure rotational Raman spectra by dual-pump CARS is discussed in detail by Lucht et al.[7] The main idea of the dual-pump CARS technique is that, by using pump lasers at two different frequencies ω_1 and ω_2, the frequency difference between pump beam 1 and the Stokes beam can be tuned into resonance with the vibrational Q-branch Raman transitions, and the frequency difference between pump beam 2 and the Stokes beam can be tuned into resonance with pure rotational Raman transitions. The energy level diagram for the simultaneous acquisition of vibrational and pure rotational spectra is presented in Fig. 1. A polarization in the medium is induced when the frequency difference $(\omega_1 - \omega_s)$ matches the resonance frequency of a vibrational Raman transition, which occurs at approximately 2330 cm^{-1}. The second

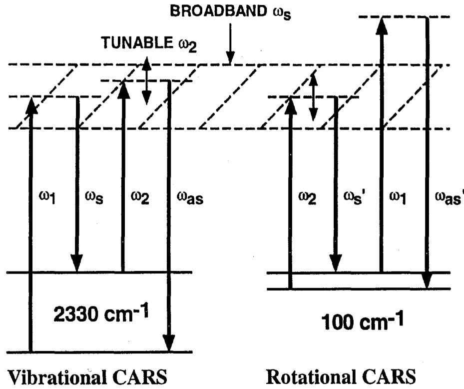


Fig. 1 Energy level schematic for simultaneous acquisition of vibrational and pure rotational nitrogen CARS spectra.

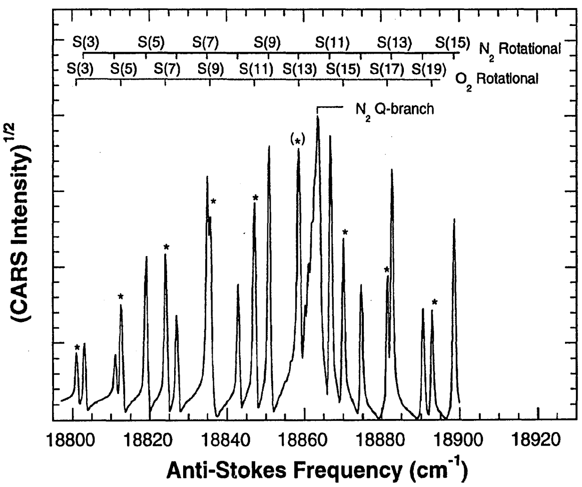


Fig. 2 Theoretical vibrational/pure rotational CARS spectra for air, P = 1 atm, T = 300 K; pure rotational transitions of oxygen are marked with asterisks.

pump beam ω_2 is then scattered from this induced polarization to produce a CARS signal at a frequency of $\omega_{as}(\text{vib}) = (\omega_1 - \omega_s) + \omega_2$. In the case of the pure rotational transitions, the roles of the pump beams ω_1 and ω_2 are reversed. A polarization is induced in the medium when the frequency difference $(\omega_2 - \omega_s')$ matches the resonance frequency of a pure rotational resonance line, which occurs at approximately 100 cm^{-1}. (In general, the Stokes laser frequency ω_s' will be slightly different than for the vibrational Raman transition, but because the Stokes laser is broadband, there will be significant spectral intensity at both ω_s and ω_s'.) Pump beam 1 will then be scattered from this induced polarization to produce a CARS signal at $\omega_{as}'(\text{rot}) = (\omega_2 - \omega_s') + \omega_1$. The anti-Stokes frequencies of the CARS signals from the vibrational and pure rotational transitions will be nearly equal and can be detected using the same digital camera. By varying the wavelength of the narrow-band dye laser, the position of the vibrational Q-branch transitions relative to the pure rotational transitions can be varied.

A representative dual-pump CARS spectrum is shown in Fig. 2. In this theoretical spectrum for air at 300 K and 1 atm, determined using the Sandia CARS code CARSFT,[17] one can see the pure rotational lines of both oxygen and nitrogen, which have similar pure rotational transition frequencies. Using a broadband laser for the Stokes beam, an approximately 100-cm^{-1} region was probed, allowing approximately a dozen pure rotational S transitions of nitrogen to be acquired. In this spectrum, the Q branch was positioned between the S(10) and S(11) nitrogen rotational lines to minimize the overlap of the Q branch with the rotational lines of oxygen and nitrogen.

The relative intensities of the vibrational Q branch and pure rotational CARS transitions are strong functions of pressure, which means that the spectral shape of the dual-pump CARS spectrum can be used to determine the gas pressure as well as the temperature. In the vibrational Q branch of nitrogen, there is substantial spectral overlap between the different vibration/rotation transitions even at 1 atm. Consequently, collisional narrowing is an important effect,[17] becoming increasingly significant at higher pressures. As pressure increases, the CARS intensity from the vibrational Q branch increases substantially because of collisional narrowing. On the other hand, at the pressure range examined in this study, the pure rotational lines are widely spaced and collisional narrowing of the pure rotational spectrum is not an important effect. For these isolated CARS transitions, the peak CARS intensity is approximately independent of pressure; consequently, as pressure increases, the relative intensities of the pure rotational lines compared to the vibrational Q-branch intensity drop substantially, as shown in Fig. 3. It is from these spectral variations that pressure is determined. As pressure increases, the rotational lines also experience pressure broadening, providing additional pressure sensitivity in the spectra. Eventually, as the rotational lines are broadened to the point that they overlap significantly, which will occur at pressures on the order of 50 atm, they will be affected by collisional narrowing. At these pressures, the dual-pump technique is not necessary for the measurement of

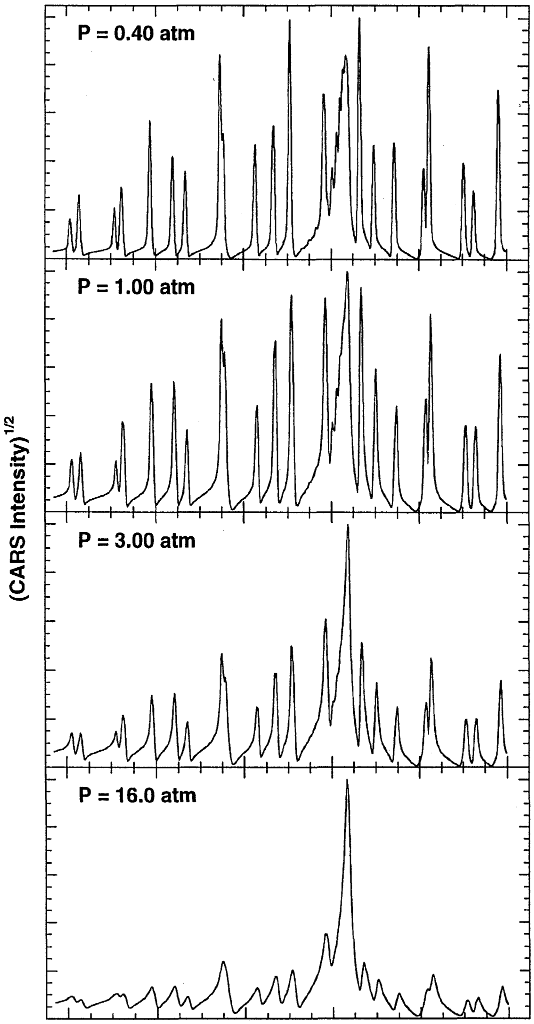


Fig. 3 Theoretical CARS spectra demonstrating the pressure sensitivity of the dual-pump CARS technique; spectra were generated using CARSFT for air at the indicated pressures and at a temperature of 300 K.

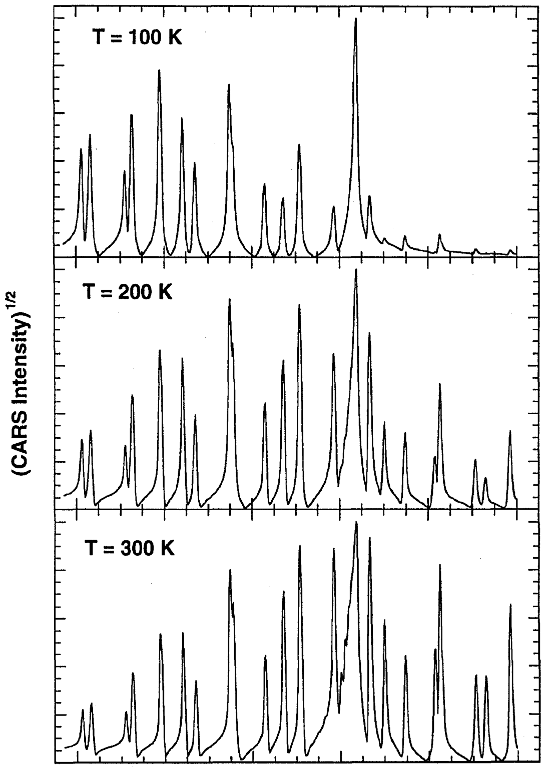


Fig. 4 Theoretical CARS spectra demonstrating the temperature sensitivity of the dual-pump CARS technique; spectra were generated using CARSFT for air at the indicated temperatures and at a pressure of 1.0 atm.

pressure, as the pressure broadening effects on the pure rotational lines' spectral shape give sufficient pressure sensitivity.

Temperature sensitivity is obtained primarily from the pure rotational CARS spectrum. As temperature decreases, the lower rotational levels become more populated, as indicated by the increased intensity of these transitions (Fig. 4). By probing numerous lines, excellent temperature sensitivity is obtained for single-shot measurements in spite of shot-to-shot fluctuations in laser intensity. With the 100-cm^{-1} range used in the temperature measurements reported in this paper, we were able to make temperature measurements across a 200-K range (100–300 K).

Data Reduction Techniques

Two data reduction methods were employed for the analysis of the experimental spectra. The time-averaged experimental spectra were fit using the CARSFT code developed at Sandia National Laboratories.[17] Single-shot spectra were fit using an integrated intensity technique.

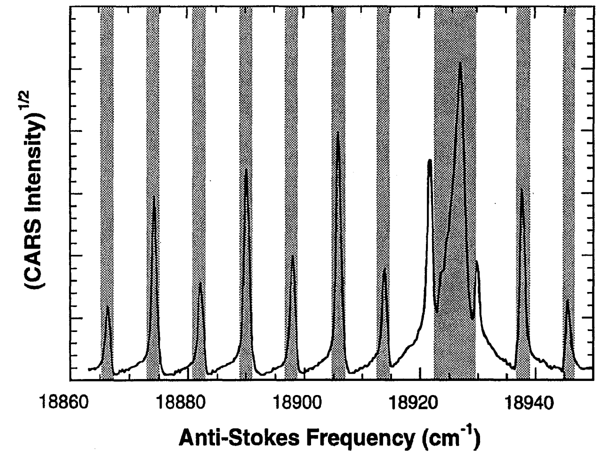
To account for collisional narrowing, the exponential gap model in the CARSFT code was used.[18] A new version of the CARSFT code was developed by Farrow at Sandia National Laboratories to correctly handle the convolution of the three laser linewidths with the molecular susceptibilities for the dual-pump CARS problem. Derivations of the dual-pump CARS signal intensity equations used in the new code can be found in Ref. 19. At pressures below 1 atm, slight differences were found between the new CARSFT code and the original code, which addressed the dual-pump CARS problem with the superposition of individually convolved vibrational and pure rotational CARS spectra. At pressures of 1 atm and above, the two CARSFT programs produced identical results.

Single-shot spectra were analyzed using an integrated intensity technique. The integrated intensities of each rotational line and the Q branch were calculated for each experimental spectrum. The integrated intensity for each pure rotational line was found by summing counts for all charge-coupled device (CCD) pixels within ± 0.75 cm^{-1} of line center. For the Q branch, the integrated intensity was found by summing counts for pixels within the section of the Q branch defined by the two bounding rotational lines, a 6.25-cm^{-1} region. Temperature information was derived from the distribution of rotational line intensities. Pressure information was derived from the ratio of the Q-branch intensity to the sum of the rotational line intensities. Thus, the spectral information is discretized as shown in Fig. 5. The experimental integrated intensities were then fit to a library of theoretical integrated intensity information developed from theoretical spectra generated using CARSFT. To calibrate the CARS system, a spectral fit was performed on a room air reference spectrum to obtain correct model parameters for the generation of the integrated intensity library.

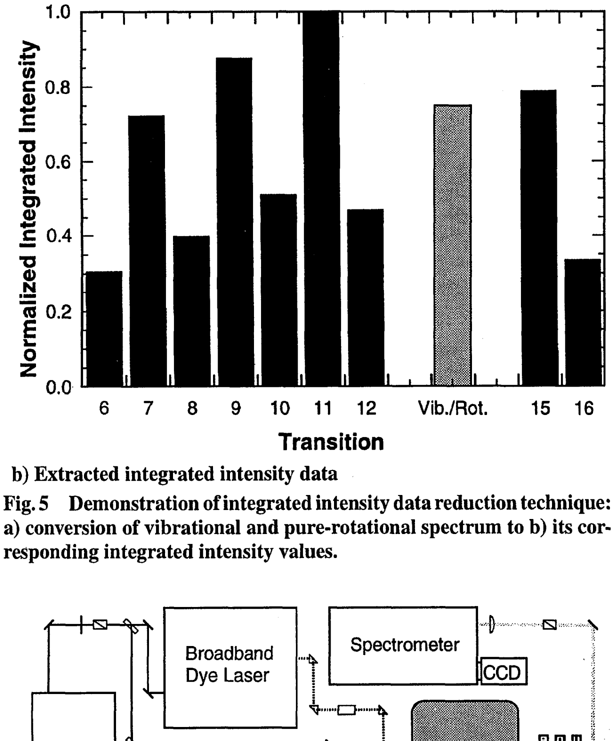
The integration reduction technique is especially useful for analysis of single-shot spectra because it averages out some camera read noise and laser mode statistical effects in the spectra. Because of this noise, the CARSFT code has a great deal of difficulty fitting single-shot spectra. Another advantage of the integrated intensity technique is a tremendous increase in data reduction efficiency over the normal CARSFT least-squares fitting technique, reducing the analysis time from over 20 min (using the dual-pump CARSFT code) to just seconds. Similar efficiency gains can also be obtained by using the CARSFT code to generate a library of theoretical spectra to which experimental spectra can be fit.

Experimental Setup

A schematic of the dual-pump CARS system used for these experiments is shown in Fig. 6. The frequency-doubled 532 nm (18,787 cm^{-1}) output of a Q-switched, injection-seeded Nd:YAG laser (Continuum NY-81C) was used as the pump beam corresponding to ω_1 in Fig. 1. The Nd:YAG laser was also used to pump a tunable narrow-band dye laser (Lumonics HyperDYE 300), which served as the second pump beam ω_2 and a broadband dye laser (Mode-X Lasers), which served as the Stokes beam ω_s. The narrow-band dye laser was tuned to 602.5 nm (16,598 cm^{-1})



a) Dual-pump CARS spectrum



b) Extracted integrated intensity data

Fig. 5 Demonstration of integrated intensity data reduction technique: a) conversion of vibrational and pure-rotational spectrum to b) its corresponding integrated intensity values.

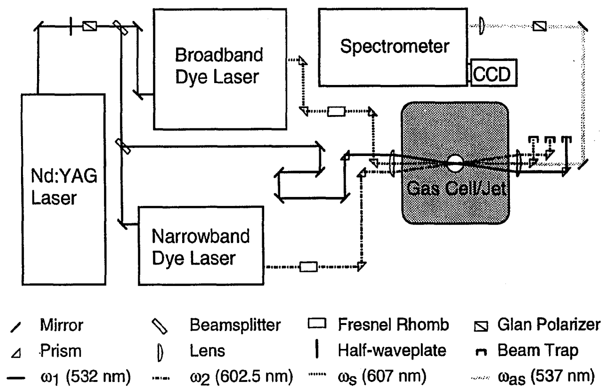


Fig. 6 Schematic of dual-pump pure-rotational/vibrational CARS system.

and had a linewidth [full width at half-maximum (FWHM)] of 0.1 cm^{-1}. The broadband dye laser radiation was centered near 607 nm (16,475 cm^{-1}) with a bandwidth (FWHM) of approximately 130 cm^{-1}. The three beams were focused into the sample medium using a folded BOXCARS arrangement. A detailed discussion of the folded BOXCARS phase matching scheme can be found in Ref. 1. The probe volume formed was approximately 50 μm in diameter and 2 mm in length. The CARS signal was dispersed using a 1-m spectrometer and imaged onto an unintensified, back-illuminated CCD camera (Photometrics). As pressure was increased, neutral density filters were added in the CARS signal channel to prevent saturation of the CCD array.

Because the frequency of the frequency-doubled Nd:YAG pump beam ω_1 is close to that of the CARS signal ω_{as}, it is difficult to eliminate scattered 532-nm light from the CARS spectrum. We used a polarization arrangement discussed in Lucht et al.[7] to reduce the 532-nm scattered light that reached the CCD detector. With respect to the polarization axis of pump beam 1, the polarization angles of the Stokes beam and pump beam 2 were 45 and 90 deg, respectively. A polarizer was placed in the CARS signal channel with its transmission axis placed at 90 deg with respect to the polarization of pump beam 1 to reject scattered 532-nm laser light. A drawback of this polarization scheme is that it reduces the intensity of the nonresonant background signal to $\frac{1}{18}$ of the case for which the polarizations of all three beams are parallel.[7] This prohibits the acquisition of nonresonant argon spectra, against which experimental CARS spectra are typically normalized to account for the spectral profile of the broadband dye laser. These dye laser spectral profiles are instead developed by referencing room-temperature, room-pressure experimental spectra to corresponding theoretical spectra. Because of the numerous pure rotational lines distributed across the dual-pump CARS spectra, the dye profile can be determined accurately. The integrated intensity reduction technique proved especially convenient for this process.

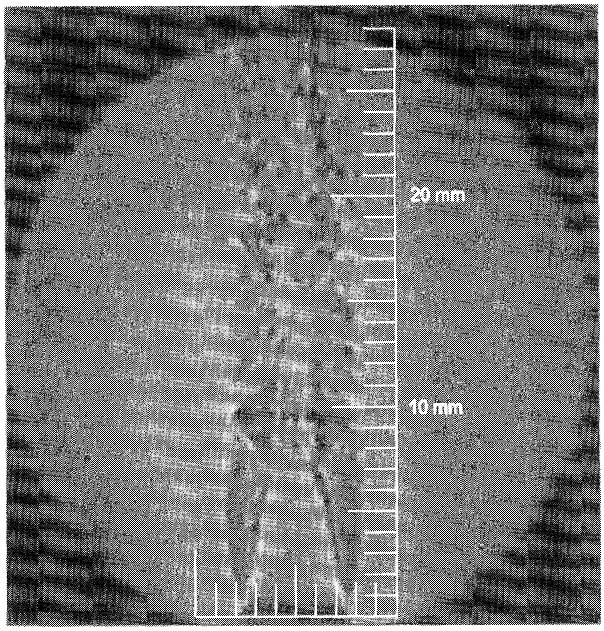


Fig. 7 Shadowgraph of underexpanded jet issuing from 5-mm-diam converging nozzle into room air; P_0 = 6.05 atm, T_0 = 295 K.

Pressure measurements at room temperature were performed in a gas cell filled with pure nitrogen at pressures ranging from 0.1 to 20 atm. In most cases, the CARS measurements were averaged over 200 shots. At pressures of 1, 2.5, 5, 10, and 18 atm, sequences of single-shot measurements were acquired to test the accuracy and precision of the technique for time-resolved measurements.

Simultaneous pressure and temperature measurements were performed in an underexpanded freejet, generated from a converging nozzle with an exit diameter of 5 mm. A shadowgraph of the jet is shown in Fig. 7. The stagnation pressure and temperature of the jet, which was exhausted into room air, were 6.05 atm and 295 K, respectively. A temperature of approximately 100 K and a pressure of approximately 0.1 atm were attained just upstream of the Mach disk, located approximately 7 mm downstream of the nozzle exit.

Results

Gas Cell

Figure 8 shows experimental CARS spectra, averaged over 200 laser shots, acquired from the nitrogen gas cell at pressures of 16.7, 1.28, and 0.30 atm; the best-fit theoretical CARS spectra are also shown. Because temperature was not varied or measured in these tests, it was unnecessary to investigate the full distribution of pure rotational lines. Thus, a narrower wave number range was examined. For the CARSFT analysis, the temperature was fixed to the experimentally measured value of 294 K. The frequency shift and dispersion, the vertical offset, the experimental intensity expansion,

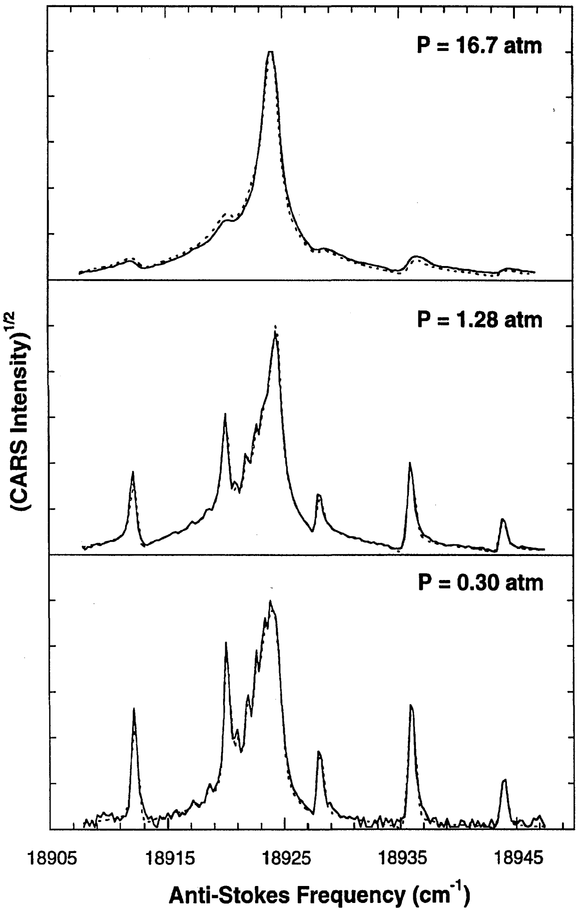


Fig. 8 Vibrational and pure rotational CARS spectra for nitrogen; experimental spectra are averaged over 200 laser shots: ——, experimental data and – – –, theoretical fit.

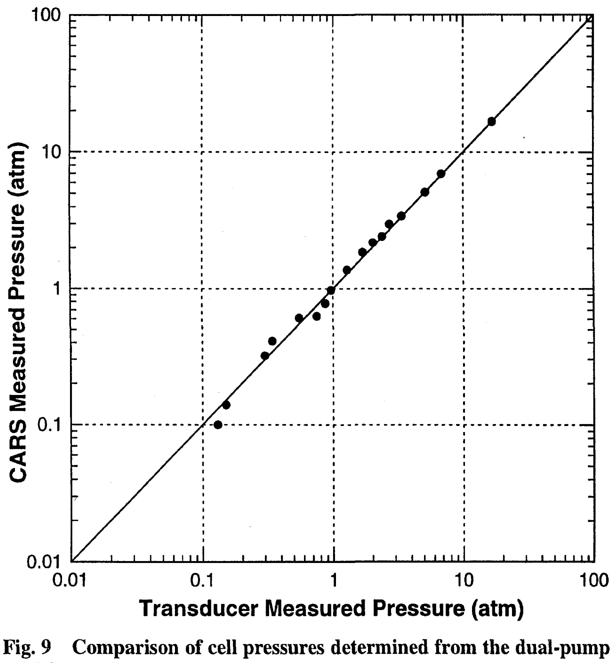
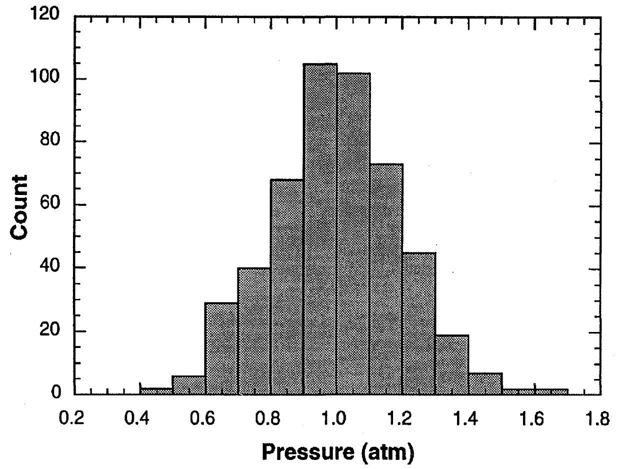
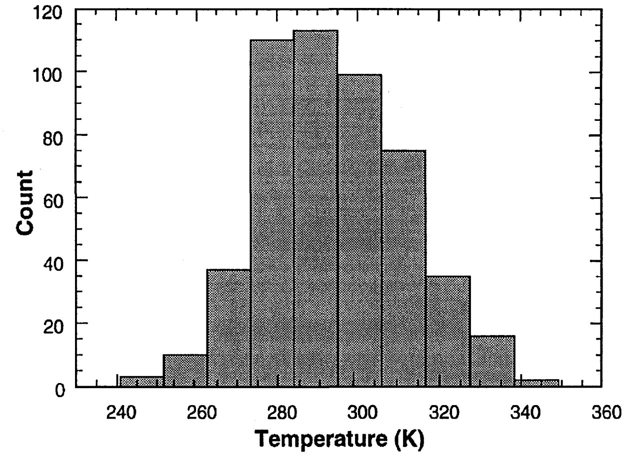


Fig. 9 Comparison of cell pressures determined from the dual-pump CARS technique and from a pressure transducer.

the transmission angle of the CARS signal channel polarizer, and the pressure were varied to achieve the best agreement between the experimental and theoretical spectra using CARSFT. A comparison between the pressures measured using the dual-pump CARS technique and those measured using a calibrated pressure transducer are shown in Fig. 9. The dual-pump CARS and transducer measurements were in good agreement. Above 0.75 atm, the mean error in the measurements was only 5%. Below 0.75 atm, error increased to an average of 15%. At low pressures there is little spectral overlap



a) Pressure



b) Temperature

Fig. 10 Measurement by integrated intensity technique for 500 room air spectra at 1.0-atm mean pressure and 294 K with 20 and 58% pressure and temperature standard deviations, respectively.

for the vibrational lines within the Q branch, and so the relative intensities of the vibrational Q-branch and pure rotational lines are fairly insensitive to pressure.

A probability density function (PDF) plot of integrated intensity pressure fits of 500 room air single-shot spectra is shown in Fig. 10a. The average of the measured single-shot pressures was within 2% of the transducer pressure. The standard deviation in the single-shot pressure measurements was approximately 20%. The limited precision of these pressure measurements is due to shot-to-shot fluctuations in the dye laser spectral profile and the relative insensitivity of the vibrational/rotational intensity ratio at pressures of 1 atm and below. At higher pressures, where collisional narrowing is more significant, the standard deviation of the pressure measurements decreases significantly, as shown in Fig. 11b. At a pressure of 3 atm, the standard deviation decreases to 10%. By 18 atm, the standard deviation is only 4%. The precision of the single-shot measurements of pressure and temperature could be improved by setting up a simultaneous reference cell system, allowing the dye laser spectral profile to be monitored on a shot-to-shot basis.[6,9]

Temperature was also determined for the room air single-shot spectra using the integrated intensity technique. The PDF of these results is shown in Fig. 10b. The average of the single-shot temperatures agrees with the measured temperature to better than 1%. Precision is much better than for the pressure measurements, with a standard deviation of less than 6%. Because numerous pure rotational transitions are being probed, temperature measurements are less affected by fluctuations in the dye laser profile than are pressure measurements.

Underexpanded Jet

Dual-pump CARS spectra were obtained along the centerline of the underexpanded jet in approximately 200-μm increments from

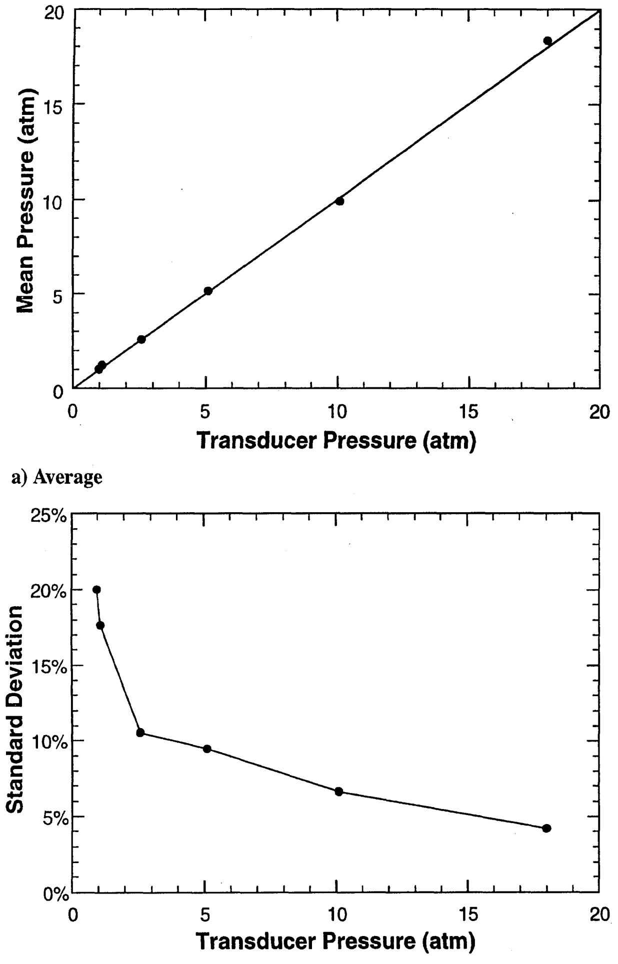


Fig. 11 Single-shot pressure measurements at various pressures; between 200 and 500 single-shot spectra were analyzed for each pressure.

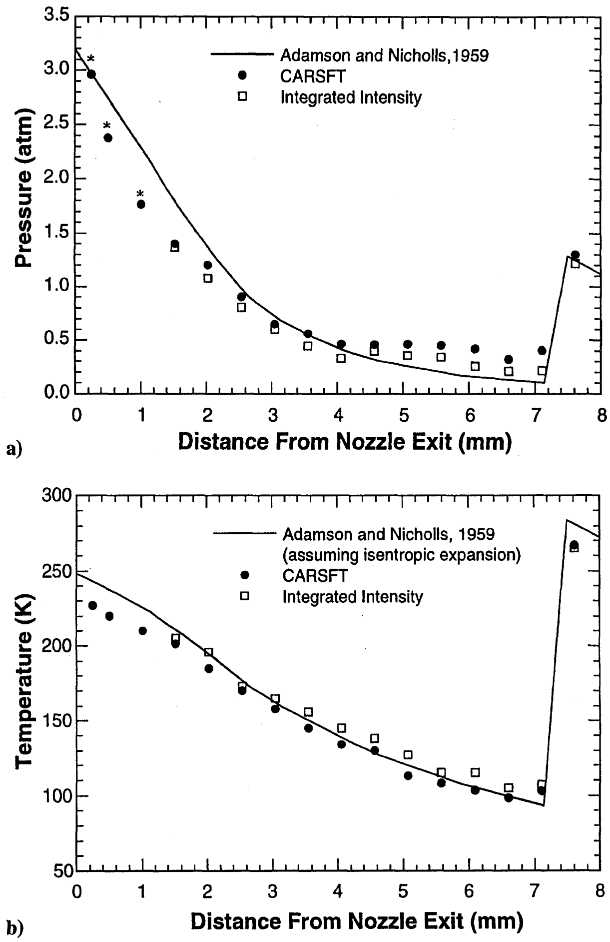


Fig. 12 Comparisons of experimentally measured pressures and temperatures to theoretical predictions for the underexpanded jet; experimental spectra averaged over 50 laser shots. Saturation of the CCD array suspected for the first three data points, marked with asterisks.

the nozzle exit to just past the Mach disk. Experimentally determined pressures were compared to a theoretical prediction found in Adamson and Nicholls,[20] which was developed using the method of characteristics. Their prediction was based on an underexpanded jet with zero backpressure. Adamson and Nicholls[20] suggest that the solution should remain applicable to any underexpanded stagnation pressure-to-backpressure ratio for a considerable distance downstream of the nozzle. The predicted temperature curve shown in Fig. 12b was generated from the predicted pressure curve in Fig. 12a by assuming an isentropic expansion. Although there is some doubt as to whether these theoretical predictions are indeed valid for the relatively low stagnation-to-backpressure ratio of our jet, the prediction from Adamson and Nicholls was the most applicable one that we were able to find for comparison to our experimental measurements.

Pressure- and temperature-fitting results, using both the CARSFT spectral fit method and the integrated intensity method, are shown in Fig. 12. Agreement of theory and experiment is very good with the exception of the lowest pressure region upstream of the Mach disk. Below a pressure of 0.4 atm the CARS spectra are relatively unaffected by pressure changes. The spectral fit method loses pressure sensitivity at approximately 0.4 atm. The integrated intensity method actually gives better fitting results, losing its pressure sensitivity around 0.25 atm. Near the nozzle exit, signal intensity is high due to the high pressures. It is suspected that insufficient neutral density filtering was used in the measurement of the first three data points, resulting in saturation of the CCD array. Temperature fits are excellent for both methods, deviating from the theoretical prediction by an average of 5%.

Spectra acquired immediately before and after the Mach disk, which are shown in Fig. 13, demonstrate well the combined effects of pressure and temperature on the vibrational and pure rotational spectra of the dual-pump CARS technique. One can clearly see the

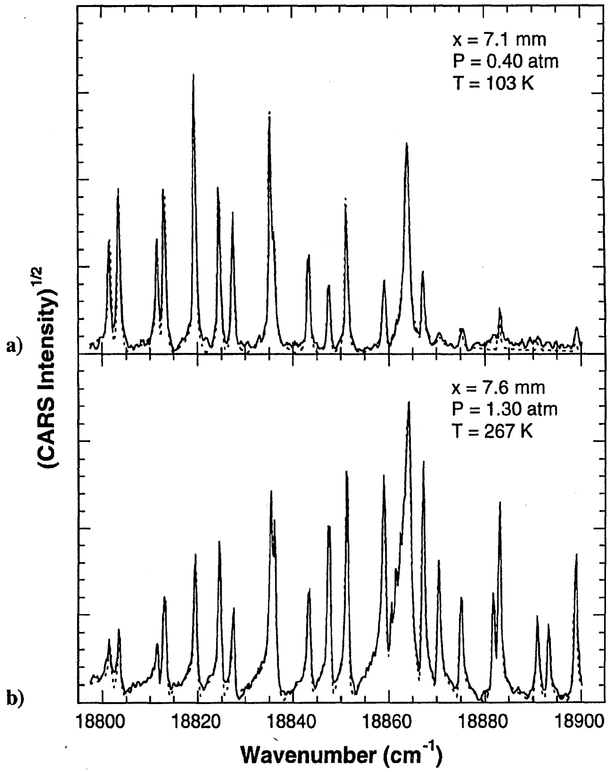


Fig. 13 Pressures and temperatures in the underexpanded jet measured by performing spectral fits using the CARSFT code. Spectra acquired a) before and b) after Mach disk.

increase in intensity of the Q branch relative to the pure rotational lines that occurs with the increase in pressure across the Mach disk. One can also see the change in the distribution of intensities of the pure rotational lines. The higher rotational level transitions become more populated at the higher temperatures in the flow downstream of the normal shock. It is interesting to note that the width of the Q branch remains relatively unchanged across the shock. This is due to the competing effects of temperature and pressure on the spectral shape of the Q branch. At the low pressure and temperature before the Mach disk, collisional narrowing effects are negligible, but the Q branch is narrowed by decreased populations of higher rotational levels within the Q branch. At the higher pressure and temperature downstream of the Mach disk, the Q branch is, on one hand, broadened due to the increased population in the higher rotational levels, but, on the other hand, is narrowed due to significant collisional narrowing effects.

Conclusions

We have developed a new CARS technique for the simultaneous measurement of temperature and pressure. The pressure measurement capability of the technique has been demonstrated in a room-temperature gas cell. Simultaneous pressure and temperature measurement capability was demonstrated in an underexpanded jet. Currently, the technique is limited to mean pressure measurements at pressures above about 0.3 atm. The technique is also useful for single-shot pressure measurements, with PDF standard deviations of 10% or less, for pressures greater than 3 atm. These experiments have demonstrated that temperature can be simultaneously measured on both time-averaged and single-shot bases with excellent results throughout a 100–300 K range.

Precision of the single-shot temperature and pressure measurements could be improved by referencing the CARS signal to a sample cell on a shot-by-shot basis. The accuracy of this technique, however, probably cannot be improved significantly, due to the minimal effects of collisional narrowing at pressures below 0.3 atm. Other CARS techniques with increased pressure sensitivity in this range will be explored.

Acknowledgments

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References

[1]Eckbreth, A. C., *Laser Diagnostics for Combustion Temperature and Species*, 2nd ed., Gordon and Breach, Amsterdam, 1996, Chap. 6.

[2]Gustafson, E. K., McDaniel, J. C., and Byer, R. L., "CARS Measurement of Velocity in a Supersonic Jet," *IEEE Journal of Quantum Electronics*, Vol. QE-17, No. 12, 1981, pp. 2258, 2259.

[3]Barrett, J. J., "Generation of Coherent Anti-Stokes Rotational Raman Radiation in Hydrogen Gas," *Applied Physics Letters*, Vol. 29, No. 11, 1976, pp. 722–724.

[4]Beattie, I. R., Gilson, T. R., and Greenhalgh, D. A., "Low Frequency Coherent Anti-Stokes Raman Spectroscopy of Air," *Nature*, Vol. 276, Nov. 1978, pp. 378, 379.

[5]Goss, L. P., Fleming, J. W., and Harvey, A. B., "Pure Rotational Coherent Anti-Stokes Raman Scattering of Simple Gases," *Optics Letters*, Vol. 5, No. 8, 1980, pp. 345–347.

[6]Murphy, D. V., and Chang, R. K., "Single-pulsed Broadband Rotational Coherent Anti-Stokes Raman-scattering Thermometry of Cold N_2 Gas," *Optics Letters*, Vol. 6, No. 5, 1981, pp. 233–235.

[7]Lucht, R. P., Palmer, R. E., and Maris, M. A., "Simultaneous Acquisition of Pure Rotational and Vibrational Nitrogen Spectra Using Three-Laser Coherent Anti-Stokes Raman Spectroscopy," *Optics Letters*, Vol. 12, No. 6, 1987, pp. 386–388.

[8]Bengtsson, P., Martinsson, L., and Aldén, M., "Combined Vibrational and Rotational CARS for Simultaneous Measurements of Temperature and Concentrations of Fuel, Oxygen, and Nitrogen," *Applied Spectroscopy*, Vol. 49, No. 2, 1995, pp. 188–192.

[9]Péalat, M., and Lefebvre, M., "Temperature Measurement by Single-Shot Dual-Line CARS in Low-Pressure Flows," *Applied Physics B*, Vol. 53, No. 1, 1991, pp. 23–29.

[10]Grisch, F., Bouchardy, P., Péalat, M., Chanetz, B., Pot, T., and Coët, M. C., "Rotational Temperature and Density Measurements in a Hypersonic Flow by Dual-Line CARS," *Applied Physics B*, Vol. 56, No. 1, 1993, pp. 14–20.

[11]Di Rosa, M. D., Chang, A. Y., and Hanson, R. K., "Continuous Wave Dye-Laser Technique for Simultaneous, Spatially Resolved Measurements of Temperature, Pressure, and Velocity of NO in an Underexpanded Free Jet," *Applied Optics*, Vol. 32, No. 21, 1993, pp. 4074–4087.

[12]Hiller, B., and Hanson, R. K., "Simultaneous Planar Measurements of Velocity and Pressure Fields in Gas Flows Using Laser-Induced Fluorescence," *Applied Optics*, Vol. 27, No. 1, 1988, pp. 33–47.

[13]Chang, A. Y., Battles, B. E., and Hanson, R. K., "Simultaneous Measurements of Velocity, Temperature, and Pressure Using Rapid CW Wavelength-Modulation Laser-Induced Fluorescence of OH," *Optics Letters*, Vol. 15, No. 12, 1990, pp. 706–708.

[14]Forkey, J. N., Finkelstein, N. D., Lempert, W. R., and Miles, R. B., "Control of Experimental Uncertainties in Filtered Rayleigh Scattering Measurements," AIAA Paper 95-0298, Jan. 1995.

[15]Elliot, G. S., and Samimy, M., "Rayleigh Scattering Technique for Simultaneous Measurements of Velocity and Thermodynamic Properties," *AIAA Journal*, Vol. 34, No. 11, 1996, pp. 2346–2352.

[16]Exton, R. J., and Hillard, M. E., "Raman Doppler Velocimetry: A Unified Approach for Measuring Molecular Flow Velocity, Temperature, and Pressure," *Applied Optics*, Vol. 25, No. 1, 1986, pp. 14–21.

[17]Palmer, R. E., "The CARSFT Code for Calculating Coherent Anti-Stokes Raman Spectra: User and Programmer Information," Sandia National Lab., SAND89-8206, Livermore, CA, Feb. 1989.

[18]Koszykowski, M. L., Farrow, R. L., and Palmer, R. E., "Calculation of Collisionally Narrowed Coherent Anti-Stokes Raman Spectroscopy Spectra," *Optics Letters*, Vol. 10, No. 10, 1985, pp. 478–480.

[19]Hancock, R. D., Schauer, F. R., Lucht, R. P., and Farrow, R. L., "Dual-Pump Coherent Anti-Stokes Raman Scattering (CARS) Measurements of Nitrogen and Oxygen in a Laminar Jet Diffusion Flame," *Applied Optics*, Vol. 36, No. 15, 1997, pp. 3217–3226.

[20]Adamson, T. C., Jr., and Nicholls, J. A., "On the Structure of Jets from Highly Underexpanded Nozzles into Still Air," *Journal of the Aerospace Sciences*, Vol. 26, No. 1, 1959, pp. 16–24.

G. Laufer
Associate Editor