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Progress in Laser Spectroscopic Techniques for Aerodynamic Measurements: An Overview

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Introduction

IN this overview, the capabilities and recent progress in laser laser spectroscopic measurement techniques for use in aerodynamic test facilities and flight research vehicles are surveyed. It also includes a compilation of the literature which is centered on this application of laser spectroscopy. The intended reader is the specialist in experimental fluid dynamics who is not intimately familiar with the physics or applications of laser spectroscopy. Thus, some discussion is also included of the nature of each laser spectroscopic technique and the practical aspects of its use for aerodynamic measurements. The specific techniques reviewed include laser absorption, laser-induced fluorescence, laser Rayleigh scattering, and laser Raman scattering including spontaneous and coherent processes.

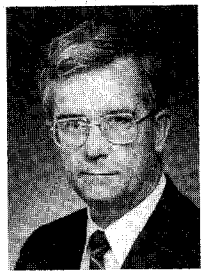
The general field of laser spectroscopic measurement technology incorporates an extensive group of approaches, many of which were originally developed to support the special needs of combustion research and the basic sciences related to it. Although the fundamental concepts that continue to evolve from those interests also find applications for aerodynamic research, a review of all of the ongoing development programs related to laser spectroscopic flow diagnostics is not practical. However, some effort is made here at least to characterize the present state of the art for applications to aerodynamic research and to identify the emerging development trends.

Throughout this overview, reference is made to "aerodynamic" research. The term is intended to differentiate between the measurement objectives of experimental aerodynamic studies in which the fluid motion is the main topic for study, and those applications that are associated with basic combustion science, plasma physics, and other sciences that tend to involve fluid dynamics more as a peripheral topic. Often, the temperatures, densities, flow speeds, and gas compositions associated with aerodynamic research are sufficiently different from those of the other applications that measurement techniques which have been developed for one are not always directly applicable to the other. Thus, by virtue of its specific requirements, laser spectroscopy for aerodynamic measurements can be considered to be a technology that is somewhat apart from other laser spectroscopic applications. On the other hand, the combustion environments which are associated with modern aeropropulsion research and development are not entirely excluded from the applications discussed here.

Attributes of Laser Spectroscopy for Aerodynamic Measurements

Generally, laser spectroscopic measurements incorporate one or more laser beams and depend on their radiative interaction with one or more spectroscopic features of the gas. Depending on the interaction process, the laser light is either absorbed or scattered by those species which are radiatively active at the wavelengths used. The intensity of the resulting radiative signal is dependent mainly on the population density of the atomic or molecular energy states which are responsible for the interaction. Measurements are thus obtained of selected population densities, from which determinations can be made of the local gas temperature and the density of the radiative species. Hence, these are the primary parameters which are determined by the methods described in this review. From them, other thermodynamic parameters such as bulk gas density and pressure can be computed using the appropriate thermodynamic relationships. Through less direct methods employing the Doppler effect or time-of-flight techniques, gas velocity may also be measured.

Because the density of specific atomic or molecular energy states generally depends on at least two independent thermodynamic variables, such as temperature and species density, the determination of any thermodynamic variables in a flow where none are known a priori requires that radiative signals from at least two independent spectral features must be measured. In some approaches, two or more spectral features are measured simultaneously using multiple detector channels or more than one laser frequency. In other approaches, spectral scanning of the laser is used to sequentially record several spectral features with a single detector channel. There are also some important special circumstances in which only the varia-



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tion of a single spectral feature is sufficient to determine the thermodynamic condition of the flow. These differences in approach are the principal features which influence the experimental complexity of each technique.

Some of the significant advantages of laser spectroscopic techniques are that they are effectively nonintrusive to the gas flow, they may be tailored to the measurement of specific thermodynamic parameters and selected species, and they can provide adequate spatial and temporal resolution for the detailed characterization of three-dimensional and time-dependent aerodynamic flows. Unfortunately, laser spectroscopic techniques can also be complex and expensive to implement, they can require optical access that is not easily provided in many aerodynamic test facilities, and they may require operation and analysis by specialists trained in fields other than fluid dynamics. Nevertheless, the significant new measurement capabilities offered by these techniques have provided compelling motivations for their use in modern aerodynamic laboratories.

Each of the following sections of this overview is devoted to one of the prevalent types of laser spectroscopic methods for flow diagnostics. Some of their applications for flow diagnostics have been reviewed recently with greater technical detail by Miles and Nosenchuck.[1] Complete descriptions of the principles, capabilities, and applications of modern laser spectroscopic methods for combustion research are given by Eckbreth.[2]

Laser Absorption

Nature of Laser Absorption

To implement laser absorption, the laser is tuned to a wavelength that is resonant with an absorbing transition of a selected species and the attenuation of the beam passing through a test region is measured. The absorption at two or more resonant wavelengths may be used to determine both the gas temperature and the density of the absorbing species. In some applications, the change in resonant wavelength owing to the Doppler effect is also measured to determine the component of flow velocity in the direction of the laser beam.

From an optical point of view, laser absorption is the least demanding diagnostic technique to implement. In its basic form, the technique requires optical access that is just large enough to allow the laser beam to pass in and out of the test region. The only significant requirement for the laser is that its wavelength match one or more spectral features of an absorbing species in the test gas. Unlike most other methods, the production of an adequately detectable signal is not dependent on the laser power, so that small, low-power lasers can be used. The minimal requirement for laser power also invites the use of fiber optics to transmit the beam which, in turn, makes the approach attractive for internal flow measurements and for flight applications.

One penalty for the simplicity of the technique is that the measurements are without spatial resolution in the direction of the laser beam. Instead, they represent integrated averages along the beam path through the test region. By increasing the optical complexity, spatial resolution can be obtained using a crossed beam, saturated-absorption technique,[3,4] but the inaccuracies of that approach which are introduced by uncertain interaction lengths and line shapes render it less favorable for obtaining spatial resolution than other completely different approaches such as laser-induced fluorescence. In the extreme, laser absorption can also be used as the basis for optical tomography to obtain complete, two-dimensional field mapping, but with the accompanying requirement for extensive optical access.[5,6]

Further requirements that accompany the use of laser absorption are imposed by the desired measurement accuracy and dynamic range. Either a sufficiently large attenuation of the laser beam passing through the test region must be obtained or methods relying on laser frequency modulation must be employed in combination with some form of heterodyne detection.[7] If laser attenuation is measured directly, the approach is usually limited to path lengths and species concentrations for which beam attenuation exceeds several percent. If laser modulation and heterodyne detection is used, attenuations much less than 1% become feasible, particularly for high modulation frequencies. Despite these potential restrictions, laser absorption techniques offer valuable new capabilities for aerodynamic measurements, as the examples to follow will show.

Aerodynamic Applications of Laser Absorption

Laser absorption spectroscopy has proven valuable as an effective diagnostic technique particularly in applications where optical access is severely limited and where the time dependence of the gas flow is of primary interest. For example, such considerations can be essential for the characterization of time-dependent flows in pulsed aerothermodynamic facilities.

In a pioneering application to a pulsed facility, Chang et al.[8] sampled time-dependent values of temperature behind incident shock waves in a shock tube containing dilute mixtures of O_2 and H_2 in argon. They used a rapidly tuned, continuous-wave (cw) ring-dye laser that repetitively scanned between two adjacent, ultraviolet absorption transitions in OH at several kilohertz. Comparisons with calculated reflected shock temperatures showed agreement within $\pm 3\%$ over the temperature range from 1300 to 3500 K.

More recently, the technique has been extended to multiparameter measurements as reported by Di Rosa et al.[9] and by Chang et al.[10] They also used a rapidly tuned ring-dye laser but scanned at 4 kHz and incorporated improved intracavity frequency doubling to obtain wavelengths near 225 nm. They demonstrated the capability to obtain simultaneous measurements of temperature, density, pressure, velocity, and mass flux behind incident shock waves, using well-separated and spectrally resolved transitions of NO. The shock tube was filled with argon containing up to 2% NO. The scalar variables were derived from an analysis of the spectral shape and relative strengths of adjacent transitions. Two laser beams crossed the shock tube at angles of 90 and 60 deg to the flow direction to allow the determination of velocity from the resonant frequency shifts owing to the Doppler effect. A similar approach was attempted based on the hot-band absorption of O_2 near 225 nm but was found to be less attractive because of the broad and overlapping spectrum of oxygen hot-band absorption at the wavelengths accessible by the scanning laser.[9]

In another study, a comparison of several cw laser absorption techniques is reported by Davidson[11] who also used absorption by OH produced behind incident shock waves in mixtures of H_2 and O_2 diluted by argon. He compared multiparameter measurements using both a scanning and a fixed frequency approach with predicted postshock values and with velocity measurements obtained from a time-of-flight technique based on H_2O photolysis. Davidson's work provides a summary of the methods and capabilities of both laser absorption techniques and shows that they both can provide postshock measurements that compare favorably with predicted values.

Laser modulation and heterodyne detection methods have also been demonstrated which allow sensitive absorption measurements for attenuations that are much less than 1%. Recent cw diode laser developments have made possible the tuning and modulation of laser frequency by coupling both ramped and sinusoidal modulation signals to the laser injection current. This approach is in contrast to earlier methods in which the laser beam was phase modulated externally using an electro-optic device.[7] Recent developments and applications of the technique to aerodynamic measurements by Philippe and Hanson[12,13] are based on a weak, near-infrared absorption band of O_2. They employed a combination of amplitude and wavelength modulation at 10 MHz, frequency scans at 10 kHz, and second harmonic homodyne detection and obtained time-

dependent measurements of temperature, pressure, density, velocity, and mass flux behind incident and reflected shock waves in oxygen. Absorption levels down to 0.2% were successfully utilized. The simple and compact optical arrangement and the low-power requirements associated with this approach may be particularly attractive for flight research applications.

Laser-Induced Fluorescence

Nature of Laser-Induced Fluorescence

Laser-induced fluorescence (LIF) arises from the same type of radiative absorption process as laser absorption but the measured signal is obtained from the subsequent spontaneous emission (fluorescence) of absorbed energy. In addition, the signal analysis must include the effects on some species of nonradiative, collisional energy transfer (quenching), which competes with the emission process as a path for energy relaxation and reduces the fluorescence signal in a manner that depends on temperature and species densities—the variables being measured.

To implement LIF, the laser beam is tuned to a suitable resonant wavelength of an absorbing species and excites a fraction of it to the upper state of a radiative transition. The excited species then spontaneously radiates the absorbed energy that is not lost to other relaxation paths, at all wavelengths that are allowed by the normal fluorescence spectrum of the excited state. Spatial resolution is obtained by observing the fluorescence from a small region of the laser beam or by imaging all of the fluorescence from a two-dimensional sheet of light. Like laser absorption, the fluorescence energy resulting from the excitation of two or more spectral features may then be used to simultaneously determine both the temperature and species density.

LIF methods can be divided into several categories, depending on the degree to which they incorporate the measurement of two or more spectral features. Some methods probe two spectral features simultaneously, but to avoid excessive experimental complexity, measurements are obtained from a small volume or "point" along a focused laser beam. The emphasis of these point measurements is usually directed toward time-resolved, simultaneous measurements of several variables.

On the other hand, if the emphasis is directed toward the acquisition of quantitative planar images of the flow, then spectral complexity is minimized in favor of image recording. This planar laser-induced fluorescence (PLIF) approach has been developed extensively and reviewed by Hanson et al.[14,15] Some recent applications of PLIF and extensions of its capabilities are described by Paul et al.[16] and by McMillin et al.[17] To implement PLIF in its simplest form, the beam from a narrowband laser is spread into a sheet which defines a plane in the flow. Quantitative images are obtained using a single camera containing a two-dimensional detector array to record and digitize the fluorescence intensity of the image field which is associated with absorption by a single spectral feature. Because only a single spectral feature is probed, the intensity field of a single PLIF image cannot generally be interpreted in terms of a specific thermodynamic variable. However, at test conditions where additional information about the flowfield is known (e.g., constant pressure), the digitized images can be processed to generate single-variable fields.

One may also argue that the determination of single thermodynamic variables from fluorescence images is not entirely necessary if the objective of PLIF measurements is to provide data for comparison with numerical code predictions of a flowfield. The fluorescence intensity depends on both temperature and species density but those variables are also coupled by the flow conservation equations. Thus, errors detected in the prediction of a single thermodynamic variable are likely to imply errors in all of the predictions, including the fluorescence intensity. The need to know which specific thermodynamic variables are in error is generally not a useful requirement in the final analysis. A numerical sensitivity analysis would define their relative errors. Thus, the fluorescence intensity could just as well be regarded as the specific variable and it could be predicted by the numerical code for comparison. The precision of any comparison of theory and experiment that is accomplished in this manner would be the same or better than one in which spectroscopic measurements were first analyzed using simplifying assumptions to somehow provide specific thermodynamic variables.

The use of LIF in either category (point or image) requires the presence of at least one suitable species which is spectroscopically simple, radiatively absorbing at wavelengths that are accessible by a laser, and thermodynamically coupled to the flow in a deterministic manner. In flows with chemical reactions, many of the reaction products provide suitable species for LIF, including a large number of chemically active, diatomic, free radicals which are formed from combinations of C, O, N, H, and others. For example, the hydroxyl radical OH has been used extensively for combustion diagnostics because it plays a significant role in the reaction chain of most hydrocarbon fuels and it is spectrally accessible using commercially available, tunable, dye lasers.

A similar situation occurs in the case of heated flows in arc jets. For example, atomic species such as copper vapor from the electrodes may be present. Marinelli et al.[18] have recently reported the use of LIF from copper atoms in arc jet flows to measure velocities based on the Doppler effect. Other preliminary diagnostics in arc jet flows, including temperature and velocity measurements based on Cu LIF, have been reported by Arepalli et al.[19]

For aerodynamic research in nonreacting flows, the choices of radiatively active species are more limited. One approach has been to seed the flow with small concentrations of a spectroscopically suitable, but chemically inactive, species. Notable examples which have been used specifically for aerodynamic measurements include sodium (Na), biacetyl ($[CH_3CO]_2$), iodine (I_2H), and nitric oxide (NO). However, whereas the use of seed materials has been successful, all of those which have been identified so far as spectroscopically suitable are either toxic, corrosive, combustible in air, or tend to condense in unheated supersonic expansions. Thus, aerodynamic testing with seed materials has been done exclusively in small facilities where the inconvenient properties of the seed species can be handled. Their use in large facilities generally does not appear to be practical.

Fortunately, the recent commercial availability of high-energy, ultraviolet, ArF excimer lasers has allowed LIF techniques to now be applied to molecular oxygen (O_2) which is naturally present in all air flows. The following paragraphs review the way in which the spectroscopy of each of these species has been used for measurements related to aerodynamic research.

Aerodynamic Applications of Laser Induced Fluorescence

Sodium

One of the first uses of LIF for aerodynamic flow diagnostics was pioneered in 1975 by Miles,[20] who proposed a means for measuring velocity based on the Doppler-shifted absorption of sodium seeded into a high-speed flow. That concept was later demonstrated by Zimmermann and Miles[21] in supersonic wind-tunnel flows of both helium and nitrogen and, finally, in a supersonic freejet of nitrogen.[22] In its final form, the technique provided two-dimensional maps of flow regions where the velocity came within a narrow range of values selected by the frequency and bandwidth of a tunable dye laser. One difficulty associated with the use of sodium in unheated flows is that it must be injected with a heated carrier gas which, in turn, can heat and perturb the primary flow. Although the approach has not been pursued recently, it may now have relevance to modern aeropropulsion research because of the presence of sodium as an impurity in rocket exhausts.

Biacetyl

Another early application of LIF for flow measurements was pioneered and reported in 1977 by Epstein,[23] who made use of the laser-induced fluorescence and phosphorescence of the organic vapor, biacetyl (2-3 butanedione). At room conditions, biacetyl is a nontoxic liquid with the relatively high vapor pressure of 40 Torr. (It is commonly used as a food additive in dairy products.) Epstein illuminated biacetyl-seeded flows of nitrogen with a two-dimensional expanded beam from a pulsed dye laser and photographed the fluorescent sheet. Since the resulting fluorescence is relatively insensitive to temperature, he was able to interpret the results in terms of density fields. Epstein applied the technique to obtain the first quantitatively recorded density fields between the moving blades of a transonic compressor rotor. His work was followed in 1979 by McKenzie et al.,[24] who monitored the time-dependent LIF from biacetyl at selected points in a supersonic, turbulent boundary-layer flow of nitrogen, using a focused cw Kr^+ laser for excitation. However, McKenzie later found that biacetyl condenses in a manner similar to water vapor so that, while it can be supercooled without condensation in a rapid expansion, it is not suitable in most flows at speeds above Mach 1 if expanded from a reservoir at room temperature. Nevertheless, subsequent work by others has shown that LIF from biacetyl has significant potential for aerodynamic research in low-speed flows where its long-lived phosphorescence can be used for velocity measurements[25,26] and its strong fluorescence can be used for the imaging of mixing flows.[27]

Iodine

Laser-induced fluorescence in flows seeded with molecular iodine has been used extensively in recent times to obtain some of the most detailed nonintrusive measurements available from complex three-dimensional flowfields. Because molecular iodine has absorption and emission spectra at visible wavelengths and a reasonably high vapor pressure (0.3 Torr at room temperature), it was an attractive candidate for early applications of LIF to flow diagnostics. Some of the first uses of molecular iodine as a seed material were for the visualization of flow structure.[28,29]

In 1982, McDaniel et al.[30] pioneered the use of I_2 LIF for quantitative measurements of density in aerodynamic flows. They showed that excitation of iodine fluorescence by a cw Ar^+ laser which is detuned from the center of a transition near the laser frequency at 514.5 nm provides fluorescence signals that are proportional mainly to density and are insensitive to the effects of collisional quenching. They used sheets of laser light to obtain photographs of density fields in the supersonic flow of a freejet of nitrogen. Subsequently, Hiller et al.[31] applied the approach to the imaging of velocity fields, using the effect of the Doppler shift on fluorescence induced by counter-propagating light sheets. As photodiode-array cameras became available, Hiller and Hanson were able to extend the technique to obtain time-averaged PLIF images of two velocity components and pressure in the shock-structured flow of a Mach 1.5 underexpanded jet.[32] Their velocity measurements relied on the comparison of sequentially recorded, digital images from four laser sheets at two frequencies and in three different directions. Their pressure measurements were based on the effects of collision broadening on an iodine absorption transition at conditions where the sensitivity to temperature is small. Measurement errors were estimated to be less than 10% for all parameters.

In more recent work, Fletcher and McDaniel[33,34] took advantage of the more complete spectroscopy allowed by point measurements with a single beam from a scanning dye laser. LIF signals from two iodine transitions were recorded including their intensity at the transition center, their Doppler shifts, and their transition widths. As a result, accurate measurements of the time-averaged temperature, density, pressure, and two velocity components were obtained without the need for any assumptions regarding the insensitivity of certain spectral features to collisional quenching or to the thermodynamic conditions of the flow. Detailed point measurements were made in the complex, three-dimensional air flow in a nonreacting, supersonic combustor with simulated fuel injection. Errors in velocity were estimated to be less than 5% while the errors in the scalar variables were approximately 2%. These comprehensive LIF data are among the first to be accepted for the validation of a modern computational fluid dynamics numerical code.[35]

Studies of the nonreacting supersonic combustor flow with injection have since been extended to incorporate advanced PLIF techniques. Hartfield et al.[36] report a method for imaging the injectant mole-fraction distribution. Planar fluorescence is induced by a broadband Ar^+ laser and collected using an intensified charge-injection-device array camera. The technique eliminates the thermodynamic dependence of the iodine fluorescence by taking the ratio of two images collected with identical thermodynamic flow conditions but with and without iodine seeding in the main flow. The results provide quantitative images of the mixing process in planes aligned with the flow. The approach was extended by Abbitt et al.[37] to obtain corresponding images in planes crosswise to the flow, thereby providing a complete three-dimensional data base of the mixing process. Most recently, Hartfield et al.[38] have incorporated the assumption that most of the flowfield satisfies the condition for which the collisional quenching rate is large compared to the spontaneous emission rate (so-called "high pressure limit"). Where this assumption is valid, the fluorescence intensity is a function of temperature and iodine mole fraction. Hartfield et al. have used this approach to add images of the temperature field to the data base. Most recently, a unified I_2 PLIF approach has been reported in which techniques for the measurement of temperature, pressure, and velocity are summarized.[39]

The I_2 LIF technique is one of the few laser spectroscopic techniques that has reached the level of development where reports of its application now dominate those of its development. On the other hand, like the preceding materials, iodine has several properties which tend to restrict its use to specialized flows in small facilities. For example, the temperature dependence of iodine vapor pressure[40] suggests that any concentration of iodine vapor will condense in a gasdynamic expansion from room temperature before reaching Mach 1. Nevertheless, the applications first described indicate that some supercooling in a freestream may be possible. In any case, I_2 LIF measurements in unheated supersonic flows should be suspect if the measurements are sensitive to the iodine concentration and they are made near cold walls, in turbulent boundary layers where flow is convected from a cold wall, or in slow expansions where time for condensation is available. Iodine is also an acute irritant and can be extremely hazardous when heated. Finally, it requires a carrier gas free of water vapor and is highly corrosive even to stainless steel. The flow facility in use by McDaniel and his colleagues is coated with Teflon to reduce corrosion. Despite these inconveniences, the compelling and well-developed measurement capabilities of I_2 LIF have been adequately demonstrated and it should continue to have many valuable applications for modern aerodynamic research in facilities designed for its use.

Nitric Oxide

To avoid the effects of condensation in unheated supersonic flows, McKenzie and Gross[41] proposed the use of nitric oxide as a seed gas in nitrogen flows. Although NO is toxic in concentrations greater than 25 ppm, the variation of its vapor pressure with temperature is similar to nitrogen. Furthermore, it is not corrosive and its two-photon absorption bands at 450 nm were accessible to the dye laser tuning range available at the time. They showed that an LIF approach based on two-photon excitation of dilute concentrations of NO could provide instantaneous temperature measurements from each laser

pulse at supersonic wind-tunnel conditions, with signal-to-noise ratios greater than 50 (i.e., less than 2% rms uncertainty). In practice, Gross and McKenzie[42] were able to achieve a minimum uncertainty of 4% which was limited by the nature of the two-photon process (see Huo et al.[43]). Subsequent advances in dye laser technology provided improved, narrowband, tunable laser light in the ultraviolet at 225 nm, which allowed NO LIF to be implemented with a more noise-free, single-photon approach. The final form of the technique allowed temperature, density, pressure, and their fluctuations to be measured simultaneously.[44] The approach was demonstrated in a Mach 2 turbulent boundary-layer flow of nitrogen seeded with 100 ppm of NO. Temperature was measured with an uncertainty of 1% rms while density and pressure were measured with an uncertainty of 2%. The instrumentation was later applied by Logan et al.[45] to evaluate the performance of a hot-wire probe in a compressible turbulent flow. She was the first to use two independent means of measuring the turbulent fluctuations of specific thermodynamic variables. As a result, she observed pressure fluctuations in the flow using LIF that were not detectable by other means and which would have introduced an ambiguity into the hot-wire probe results.

Nitric oxide has also been used as the fluorescent species in PLIF imaging of aerodynamic flows. In one example, Paul et al.[46] obtained velocity field images in a Mach 7 underexpanded freejet of nitrogen with 0.5% NO. Their unique approach was similar to that described previously for Na and I_2 LIF, except that broadband laser excitation was used. The forward and backward Doppler shifts of a resonant NO transition were indicated by the change in their convolution with the fixed, broadband, laser spectral distribution. In other PLIF work, McMillin et al.[47] used NO LIF to obtain images from the flow behind incident and reflected shock waves in a shock tube and from the shock structure and flow over a two-dimensional wedge. Their excitation scheme allowed them to obtain quantitative images showing the effects of vibrational relaxation.

Another recent application of NO LIF is reported by Di Rosa et al.[48] who use it in conjunction with a rapidly scanned, cw ring-dye laser to obtain simultaneous measurements of rotational temperature, pressure, and velocity in an underexpanded freejet of nitrogen that was seeded with 0.5% NO. The technique is an extension of related approaches based on laser absorption[9,10] but it benefits from the spatial resolution provided by LIF. The rotational temperature measurements reveal that at high Mach numbers, a systematic departure from the isentropic temperature was present, as might be expected in a rapidly expanding flow with thermodynamic nonequilibrium.

Although NO LIF successfully provides new measurement capabilities that have been demonstrated for compressible and time-dependent flow diagnostics, it suffers from several drawbacks that limit its broader use for aerodynamic research. One is the fact that NO is a toxic hazard for short-term exposures to concentrations greater than 100 ppm. Another is that nitrogen is required as the diluent, rather than air, because NO emission is severely quenched by oxygen. Both of these considerations tend to restrict the use of flows seeded with NO to small facilities which are designed specifically to handle gases other than air. On the other hand, NO is free of condensation in unheated flows up to Mach 3 and it can be produced thermochemically in high-enthalpy, aerodynamic flows and in combustion flows of current interest. However, when NO is produced thermochemically, oxygen and other efficient quenching species are also likely to be present in quantities that will complicate its use as a means of measuring temperature. Nevertheless, its use for measuring molecular velocities and the use of its concentration as a reaction process indicator should remain as noteworthy diagnostic possibilities for which a great deal of development has already been completed.

Oxygen

The development of commercial ArF excimer lasers, which operate with high pulse energy over a narrow band centered at 193 nm, has recently motivated several investigations of LIF from molecular oxygen. The tuning range of the laser encompasses several rotational-vibrational transitions within the ultraviolet absorption spectrum of O_2. As a result, thermodynamic measurements based on O_2 fluorescence in air have become possible without the need for seed materials.

The O_2 LIF concept was first explored by Massey and Lemon[49] who used a home-built, tunable, ArF laser to evaluate the feasibility of the concept for measuring temperature and density in air. Their work was followed by others in several laboratories using commercial lasers. For example, Lee and Hanson[50] calculated the O_2 absorption and fluorescence related to excitation by a broadband ArF laser to evaluate the feasibility of applications to combustion and heated air flows. The predicted fluorescent yields were large enough to encourage the continued investigation of O_2 LIF. Subsequently, Lee et al.[51] demonstrated PLIF images of the temperature field in an air flow heated by an electric torch. The measured fluorescence signal was converted to temperature using calculations of its temperature dependence and reliance on the fact that the pressure and oxygen mole fraction in the flow were constant. Cohen et al.[52] reported a similar demonstration of O_2 PLIF to obtain fluorescence images from a supersonic jet of heated air.

In another approach, Gross and McKenzie[53] performed calculations of the O_2 fluorescence intensity for conditions related to hypersonic wind-tunnel flows at low temperatures. They predicted signal strengths that were sufficient to provide simultaneous point measurements of temperature, density, and their fluctuations in air flows at temperatures below 500 K. Their approach relies on the use of a single, narrowband, tunable, ArF laser to provide both the broadband fluorescence from O_2 and the Raman scattering from O_2 and N_2 from each laser pulse. A similar approach based on the use of O_2 LIF in combination with Rayleigh scattering was proposed by Miles et al.[54] to obtain two-dimensional images of temperature and density fields. The stronger signals from Rayleigh scattering, as compared to Raman scattering, make two-dimensional imaging more feasible. In either approach, when the flow is nonreacting, the fluorescence signal is sensitive to both temperature and density whereas the Raman or Rayleigh signal depends only on density. The simultaneous measurement of both signals then allows the simultaneous determination of temperature and density. Moreover, for flows in thermochemical equilibrium, an equation of state can also be defined from which pressure can be calculated.

The capabilities of the O_2 LIF/Raman technique were initially characterized experimentally by Laufer et al.[55] who identified spectral regions in which nonlinear, multiphoton processes interfere with the measurement. They also showed that sufficiently large signal-to-noise ratios can be obtained in the flow of a typical Mach 10 wind tunnel to allow measurements of temperature and density with uncertainties of 2% or less, from a single laser pulse. Fletcher and McKenzie[56] further defined some limitations of the technique which are caused by multiphoton processes and they demonstrated the acquisition of simultaneous temperature and density measurements in a flowing air cell at conditions which approach those in a hypersonic wind tunnel. Most recently, the technique has been demonstrated in the same fully turbulent boundary-layer flow at Mach 2 that was used previously to demonstrate the NO-LIF method. Simultaneous measurements of temperature, density, pressure, and their fluctuations owing to turbulence were obtained which are essentially identical to the NO-LIF results.[57] The technique now appears to be fully capable of providing sufficiently accurate measurements in air flows at temperatures down to 60 K and densities down to 0.01 Amagat. These conditions match those in the freestream of a hypersonic wind tunnel at Mach 10 for stagnation conditions of 100 atm and 1000 K.

Molecular Velocimetry Using Flow Tagging and Laser-Induced Fluorescence

The applications of LIF already described have included methods for determining flow velocity, but they all have relied

on some form of the Doppler effect. Because the use of the Doppler effect depends on measurements of the frequency shift of a single spectral feature, such techniques generally require measurements that are sensitive to small frequency changes and sufficiently high flow speeds in the direction of the probing laser beam to accurately determine velocity. Moreover, by the nature of their implementation, many of the Doppler methods provide only time-averaged measurements.

In contrast, approaches based on "flow tagging" depend on some kind of laser-induced interaction with the flowing gas to spectroscopically mark a small volume of the flow, followed by LIF to locate its convected distance downstream in a specified time interval. If the marking can be done without disturbing the local gasdynamic conditions, velocities of almost any magnitude and direction can be measured accurately and with fewer demands on the spectroscopic quality of the laser system than required by Doppler-based methods. Furthermore, most flow tagging methods provide local, instantaneous velocities along one or more lines. The results can then be analyzed to obtain average velocities, fluctuation statistics, spatial correlations, and vorticity.

In one pioneering approach to flow tagging, Boedeker[58] used a KrF ultraviolet laser operating at 248 nm to photodissociate H_2O which was seeded into a supersonic freejet of nitrogen. An unfocused dye laser at 308 nm provided PLIF images of the downstream location of the photofragment OH. The technique has particular utility for velocity measurements in combustor or rocket exhausts where water vapor is present naturally as a combustion product. A recent application of the technique is reported by Goss et al.[59] and its application to the exhaust flow of the Space Shuttle main engine and to general flows containing water vapor is evaluated by Boedeker.[60] A related approach based on the photolysis of hydrocarbon species to yield C, C_2, and CN in high-speed combustion flows is described by Goss et al.[61]

Using a much different spectroscopic interaction, Miles et al.[62] have pioneered a method of flow tagging that is suitable for general applications to aerodynamic research in a wide variety of air flows. In its original form, their technique is based on vibrational tagging of O_2 using a frequency-doubled Nd:YAG laser at 532 nm and a dye laser at 580 nm to generate two focused beams with a frequency difference equal to the vibrational Raman frequency of O_2. The resulting stimulated Raman process elevates a substantial fraction of the O_2 molecules in the two overlapping beams to the long-lived, first excited vibrational state, thereby marking a region of the flow. The excited oxygen molecules are then located downstream using PLIF images generated with a narrowband ArF laser at wavelengths tuned to induce fluorescence only from the first vibrational state. The technique is now called "Raman excitation plus laser-induced electronic fluorescence" (RELIEF). Recent developments are reported by Lempert et al.[63] in which the dye laser has been replaced with an O_2-He filled Raman frequency converter[64] that automatically converts part of the Nd:YAG beam to the required frequency shifted by the Raman vibrational transition in O_2. As a result, the complexity and cost of the apparatus are reduced and its operation is greatly simplified.

RELIEF has been applied by Miles and co-workers to the study of a free shear layer in an axisymmetric jet of air at Mach 3.6 and at subsonic speeds.[65] Using single lines written in the Mach 3.6 flow, velocities from 100 to 600 m/s were measured with an uncertainty of $\pm$ 20 m/s. The complete data set includes average velocity distributions, their fluctuations, and their spatial correlations in the regions upstream and downstream of the Mach disk. Two lines in the flow were tagged simultaneously so that both streamwise and transverse velocity correlations were also obtained. The measurements appear to be of sufficient quality to provide meaningful data for comparison with numerical simulations.

More recently, the technique has been applied to the observation of small-scale velocity structures in high Reynolds number turbulent flows.[66] Lines which are approximately 100 μm thick and 1 cm long were written into the flow of a turbulent freejet and observed with a spatial resolution on the order of tens of micrometers. The data lead to measurements of the turbulence intensity, the lateral correlation function, and the Taylor microscale. The second-order structure function at one location in the flow was shown to have a slope of two-thirds in the linear portion of a logarithmic plot, in accordance with the expected Kolmogoroff energy scaling relationship. This is the first demonstration of instantaneous turbulence measurements with microscale spatial resolution.

As noted by Miles, the RELIEF approach is presently limited by the thermal population of the first excited vibrational state to temperatures below 800 K and by the minimum detectable signal to densities above 0.1 Amagat. In addition, the vibrational relaxation time limits the maximum observation time in pure air to 100 μs or less. Thus, if a maximum convection distance of 1 mm is specified, the corresponding minimum velocity is 10 m/s. Nevertheless, these limitations suggest that RELIEF is well suited to the diagnostics of most subsonic flows and nearly all unheated, supersonic flows.

Rayleigh Scattering

Nature of Laser Rayleigh Scattering

Laser Rayleigh scattering may be described as an elastic, radiative interaction with the gas in which light from a laser beam is scattered at nearly the same wavelength as the laser. The scattered light intensity is proportional primarily to the sum of densities of all species in the gas, weighted by their respective Rayleigh cross sections. Spatial resolution is obtained by observing the scattered light from a small region of the laser beam or by imaging the scattering from a two-dimensional light sheet.

From a spectroscopic point of view, Rayleigh scattering is the least demanding method to apply. It does not rely on any spectral resonances or seed material, all species in the gas scatter the laser beam, and the laser can be at any wavelength. A particularly noteworthy feature is that the scattering intensity is inversely proportional to the fourth power of the laser wavelength, which encourages the use of short-wavelength lasers. Consequently, the recent commercial availability of high-energy, ultraviolet (uv), excimer lasers and the concurrent availability of uv-sensitive photodetector arrays have allowed new advances to be made in the use of Rayleigh scattering as a viable technique for obtaining instantaneous, quantitative, two-dimensional density images from a high-speed flow.

On the other hand, a discouraging feature of Rayleigh scattering is the lack of spectral difference between the light scattered from the gas and background light scattered by any other object that can illuminate the field of view. Facility walls and particulates in the flow are commonly encountered sources of background scattering. In addition, internal scattering by the dense material of any window or optical element in the laser beam path causes intense light to propagate in all directions within the optical element and to reflect and scatter from its surfaces, edges, and supports. Moreover, the scattering is usually greatest in the forward direction so that intense scattered light from the laser travels unfocused in the direction of the beam. Consequently, the reduction of background light to levels below that scattered by the gas flow is often difficult. Nevertheless, methods for achieving adequate background reduction have been developed and successful measurements of gas density have been performed with meaningful spatial and temporal resolution.

Aerodynamic Applications of Laser Rayleigh Scattering

In applications done prior to the availability of ultraviolet lasers, the spectral simplicity of Rayleigh scattering still attracted its use for aerodynamic measurements. The technique was applied using a variety of cw and pulsed lasers operated

mostly at visible wavelengths. For example, one of the early uses of laser Rayleigh scattering was to characterize the onset of condensation in supersonic flows. Since molecular clusters which are formed by condensation scatter light in proportion to their size, the onset of condensation and the subsequent cluster growth can be observed as an enhancement of the Rayleigh signal. Measurements based on that approach were reported in 1973 by Lewis and Williams,[67] and later by Williams and Lewis,[68] who used a cw Ar^+ laser for measurements in a freejet expansion of argon to detect the Ar_2 dimer formation and cluster growth. Recent studies using high-energy pulsed lasers have applied Rayleigh scattering to air flows in supersonic wind tunnels and found the presence of previously unsuspected aerosols, apparently produced by oxygen condensation.[69,70] Although the aerosol presence would not be expected to compromise the aerodynamic quality of the test facility, it does jeopardize the usefulness of Rayleigh scattering as a means of accurately measuring density in those facilities.

In a much different approach, Cattolica et al.[71] analyzed the spectral shape of Doppler-broadened Rayleigh scattering to determine the temperature of heated gas flows. Measurements in air, nitrogen, oxygen, and argon were obtained which established the criterion for estimating the minimum densities in those gases at which Brillouin scattering—an interaction between a radiative field and acoustic waves—begins to influence the spectral shape of the scattered light. The intended applications of this approach were to combustion environments where the use of Rayleigh scattering could be extended to measure temperature as well as density.

Examples of other aerodynamic applications of laser Rayleigh scattering with visible-wavelength lasers include the measurements by Hoppe and Honaker[72] of time-averaged freestream densities in hypersonic helium flows and the spatially and temporally resolved measurements by Pitts and Kashiwagi[73] of the statistics of turbulent mixing in a methane jet issuing into a slow air flow.

Rayleigh scattering was also used by Escoda and Long[74] for one of the first examples of quantitative, two-dimensional imaging of a gas flow. Images of the scattered light were recorded with a low-light-level television camera to provide instantaneous concentration fields in a coflowing turbulent jet of freon and air. The technique was shown to have adequate spatial resolution to measure the effects of molecular diffusion at the boundaries of turbulent eddies.

Simultaneous measurements of density and velocity have been accomplished by combining laser Rayleigh scattering from the gas with laser Doppler velocimetry (LDV) using particle scattering. The principal difficulty in this approach is caused by the opposing requirements of the velocity measurements, which require particulates in the flow, and the density measurements, which are contaminated by particle scattering. Proper triggering and time gating of both signals to separate them temporally allowed nearly simultaneous measurements to be obtained without conflict. In a series of such experiments, Driscoll et al.[75] and Schefer and Dibble[76] measured the velocity-density correlation statistics in a turbulent nonpremixed flame at high Reynolds numbers. In another application, de Groot et al.[77,78] obtained simultaneous measurements of density and velocity in a cold air flow over a backward-facing step with injection of CO_2. Their flow configuration simulated the complex turbulent flow in a solid fueled ramjet with mass injection from the burning solid fuel.

More recently, the commercial availability of ultraviolet ArF excimer lasers operating at 193 nm has facilitated significant new advances in the use of Rayleigh scattering for aerodynamic measurements. The uv scattered light at 193 nm is 60 times more intense that it would be for the same laser intensity at the visible wavelength of 532 nm, while the reflective efficiency of solid materials contributing to the background is significantly less. These features, coupled with the availability of uv-sensitive, intensified, detector-array cameras, has made the acquisition of density field images much more practical. Using the advanced instrumentation, Miles and Lempert[70] and Smith et al.[79] have successfully obtained instantaneous images of a turbulent boundary layer, including its eddy structure, in a Mach 2.5 flow. The digitized images were also processed to reduce the background and to generate spatial correlation contour maps of the density field. Similar images were obtained of the shock wave/boundary-layer interaction region at the foot of a Mach 2.5 flow over a 16-deg wedge.[79] References 65 and 70 also demonstrated the combination of the Rayleigh technique with simultaneous velocity measurements using RELIEF.

In addition, Miles and Lempert[70,80] and Miles[81] have demonstrated a filtered Rayleigh technique, in which an iodine-vapor filter is used to selectively separate the Rayleigh-scattered light which is spectrally shifted by the Doppler effect from the unshifted background light. The technique significantly reduces the background and provides images of the density field for velocities in a range selected by the filter.

Ultraviolet laser Rayleigh scattering has also been studied in detail for a flight application. McKenzie[82] showed that it provides an unambiguous means of measuring the highly variable, upper stratosphere density that is encountered by the Space Shuttle during entry. Such measurements are needed for analysis of the entry flight dynamics and the 193-nm radiation from an ArF excimer laser is ideal for the task. A beam from a laser located onboard the Shuttle can propagate out through the crew hatch window in a direction normal to the flight direction and the backscattered Rayleigh light can be collected through the same window. With laser performance equal to that available from commercial systems, atmospheric densities can be measured with less than 1% uncertainty and a spatial resolution of 1 km along the flight path, over the altitude range of 50–90 km. Aerosols in the atmosphere limit the accuracy of the technique at lower altitudes. Experimental verification of the signal linearity and the expected signal-to-noise ratios were demonstrated in a simulation facility at conditions duplicating the signal levels of the flight environment.

The results also point to the unique capability of the Space Shuttle as a hypersonic test facility for using laser spectroscopic techniques. It is the only vehicle in existence with adequate size and onboard utilities to support laser spectroscopic measurements using laboratory-size lasers. Moreover, the Shuttle hypersonic flight conditions in the upper atmosphere create significant, nonequilibrium thermochemical effects in the shock layer that are similar to those for other advanced vehicles and are not duplicated by any ground-based test facility.

Raman Scattering

Nature of Laser Raman Scattering

Raman scattering may be described as a radiative interaction in which light from one or more laser beams is scattered inelastically by the gas at wavelengths shifted from that of the laser. The wavelength shifts depend on the structure of the scattering species and make up a spectrum representing all of the Raman resonances within the species of the test gas which are promoted by the Raman process. The spectral intensity distribution may then be used to determine the temperature and species densities.

Like Rayleigh scattering, Raman scattering also can be implemented using one laser with an arbitrary wavelength. This feature allows the laser selection to be based on the laser attributes rather than those of the gas species being probed. Also like Rayleigh scattering, the intensity of Raman scattering is inversely proportional to the fourth power of the scattered light wavelength, which again encourages the use of short-wavelength lasers. Typically, repetitively pulsed lasers are used because they can deliver the highest energy or average power to a small sample volume. (When only time-averaged measurements are required, the critical laser performance pa-

rameter is always time-averaged power, even for pulsed lasers. When single-pulse measurements are required, the laser pulse energy is the critical laser parameter.) However, unlike Rayleigh scattering, the Raman signals are at wavelengths different from the laser wavelength and consequently are affected very little by background scattering. In addition, the Raman effect is an interaction which is driven by the radiative field and, consequently, is not affected by collisional quenching. Hence, the only collisional processes that must be considered in the analysis of Raman signals are those which affect the spectral widths of the Raman transitions.

Raman scattering techniques may be divided into two categories: those relying on *spontaneous* scattering and those which may be regarded as *coherent* interactions. Each has advantages in different test environments so that the selection of a measurement approach is usually motivated by the conditions at which the measurements must be made.

Spontaneous Raman Scattering

One feature of spontaneous Raman scattering is that light from a single laser beam is weakly scattered in all directions. Spatial resolution is obtained by observing the scattered light from a small region of the laser beam. In addition, the intensity of scattered light varies linearly with the intensity of the incident laser light, thereby allowing the Raman signals to be easily normalized by the pulse-to-pulse variations in laser energy. The spectral analysis of spontaneous Raman scattering is relatively straightforward because the contributions from overlapping spectral features can be linearly summed. For simple diatomic molecules, the only uncertainty which may remain is the magnitude and temperature dependence of collisional broadening effects. However, most spontaneous Raman measurements for aerodynamic applications depend on the spectrally integrated energies in Raman transitions, rather than their spectral shape.

On the other hand, the efficiency of spontaneous Raman scattering at visible wavelengths is ordinarily very weak (approximately 1000 times less than Rayleigh scattering). Furthermore, since the scattering occurs in all directions, only a fraction of the signal can be collected. In principle, the contributions from each species can be discriminated in the Raman spectra, but in practice, only scattering from the major species can be detected with useful accuracy because of the weak Raman signal. For flows with combustion, interference from soot incandescence and background luminescence can render the approach impractical,[2] except for applications to certain clean environments such as hydrogen-air flames. For aerodynamic measurements, which are often made in low-density flows, the weak signals at visible wavelengths usually require photon-counting detection while the associated photon-statistical noise requires temporal averaging to achieve adequate measurement accuracy.

Now that high-energy uv lasers are commercially available, the utility of spontaneous Raman scattering is much greater because advantage may be taken of the large increases in Raman scattering intensity at uv wavelengths. For example, when only the increase associated with the fourth power of scattered light wavelength is considered and a comparison is made of the Raman intensity from an ArF excimer laser operated at 193 nm with that from a frequency-doubled Nd:YAG laser at 532 nm, the ultraviolet scattering should be at least 72 times greater for O_2 (1555 cm^{-1} vibrational Raman frequency shift) and 82 times greater for N_2 (2331 cm^{-1}). Furthermore, Bischel and Black[83] measured the effects of electronic resonance enhancement of the Raman cross sections for several important molecules as a function of wavelength and found that N_2, H_2, D_2, and CH_4 have an additional enhancement of 2–4 at 193 nm, depending on the molecule. None of them have electronic resonances which are close to 193 nm. However, O_2, which has an electronic band that encompasses 193 nm, has a very significant electronic resonance enhancement factor of 27. The combined effects of these Raman enhancements, coupled with the high pulse energies available from uv excimer lasers, has led to a renewed interest in spontaneous Raman scattering as a viable measurement technique for both combustion and aerodynamic research.

Applications of Spontaneous Raman Scattering

Despite the difficulties associated with weak signals at visible wavelengths, numerous applications of spontaneous Raman scattering have been demonstrated in aerodynamic facilities in the past 20 years using lasers at visible wavelengths. In the period up to 1980, pulsed, flashlamp-pumped, ruby and dye lasers, and high power, cw Ar^+ lasers were the primary choices available. Techniques and applications of spontaneous Raman scattering using those lasers were reviewed in 1977 by Lederman.[84] He included a detailed description of the spectroscopy and reviewed its application to the measurement of temperature and density in flowfields. A similar description and a more recent review of related applications to combustion environments is provided by Eckbreth.[2]

An example of an early aerodynamic application was reported in 1974 by Hillard et al.,[85] who demonstrated the first simultaneous measurements of time-averaged temperature, density, and velocity using spontaneous Raman scattering combined with LDV. Their measurements were made in the flowfield over a flat-plate model in a Mach 5 wind tunnel. They used cw Ar^+ lasers at two wavelengths to separate the LDV velocity measurements from the scalar variables and they used photon-counting detection to record the Raman signals. They demonstrated that Raman measurements could be obtained without excessive background interference from the LDV particles in the flow.

When flashlamp-pumped dye lasers with pulse energies exceeding 1 J became more reliable, single-pulse spontaneous Raman scattering measurements and their combination with laser velocimetry became more viable. This improved capability allowed the simultaneous measurements of temperature and species densities, their fluctuations, and their correlations.[86] The technique was applied to H_2-air turbulent diffusion flames by Drake et al.,[87] who studied the probability distribution functions and correlations of the scalar variables (temperature and species mole fractions), and by Dibble et al.,[88] who extended previous work using Rayleigh scattering combined with LDV[75,76] to obtain improved measurements of velocity-scalar correlations. Quantitative, two-dimensional imaging using Raman scattering was also demonstrated by Long et al.,[89] who extended previous work using Rayleigh scattering[74] to obtain two-dimensional maps of the instantaneous, CH_4 concentrations in a turbulent, nonreacting, CH_4 jet issuing into air.

More recent applications of spontaneous Raman scattering take advantage of the commercial availability of tunable, high-energy, ultraviolet excimer lasers and the greatly enhanced Raman signals that they provide. In one example, Pitz et al.[90] used a broadband KrF laser operating at 248 nm to evaluate the interference of LIF which underlies the Raman vibrational spectra of H_2, H_2O, and O_2 in hydrogen-air flames. They found that interfering LIF from OH and O_2 diminishes the usefulness of the technique with a broadband laser. However, in related work using a narrowband, tunable KrF laser, Shirley,[91] and later Wehrmeyer et al.[92] and Cheng et al.,[93] showed that wavelengths within the tuning range of the KrF laser can be found where the interference from LIF is greatly reduced and the Raman spectra of N_2, O_2, H_2O, and H_2 can be observed with sufficient accuracy to allow single-pulse measurements of temperature and major species concentrations. In an aeropropulsion application, Cheng et al.[94] report experiments in which spontaneous Raman scattering from a narrowband laser is combined with OH LIF to obtain simultaneous point measurements of temperature, mixture fraction, and multispecies concentrations in the reacting flow of a Mach 2 burner. More recently, Shirley[95] has applied a narrowband KrF excimer to obtain nitrogen Raman spectra in

a gas turbine combustor test rig burning methane or Jet A fuel.

A similar approach has been taken by Laufer et al.[55] and subsequently by Fletcher and McKenzie[56,57] in the application of spontaneous Raman scattering for the single-pulse measurement of density in hypersonic air flows. As described in the Aerodynamic Applications of Laser Induced Fluorescence section, an ArF laser operating at 193 nm was tuned to an O_2 transition for LIF while the Raman scattering by O_2 and N_2 from the same beam was used to provide measurements of density. The density measurements are possible with an uncertainty of 2% for a Mach 10 freestream density of 0.01 Amagat because of the large electronic resonance enhancement of the O_2 Raman cross section at 193 nm. Fletcher and McKenzie[56] also show that fluorescence from multiphoton processes, which become dominant at high laser fluence (pulse energy per unit area), can interfere with the Raman measurement when the laser is tuned to certain LIF transitions.

Coherent Raman Scattering

Coherent Raman processes generally involve the interaction of the gas with light from several laser beams having selected wavelength differences. Since only the wavelength differences are required to be resonant with a molecular transition, the absolute wavelength of one laser may again be chosen arbitrarily. As with spontaneous Raman scattering, coherent Raman signals would also be increased by selecting ultraviolet lasers but the advantage of that approach is offset in this case by the advantages gained with well-behaved lasers operating at visible wavelengths. Compared to uv lasers, visible lasers offer beams with better spectral quality, a greater range of tunability, equally high pulse energy, and the ability to transmit light through the conventional glass windows which are found in most aerodynamic facilities.

A variety of coherent Raman techniques have been demonstrated for spectroscopic and nonlinear optical applications, but not all appear to be practical for flow measurements. A review by She et al.[96] discusses those techniques which appeared to offer some potential for practical application. Of the techniques considered, stimulated Raman spectroscopy (SRS) and coherent anti-Stokes Raman spectroscopy (CARS) have emerged as the methods which have been demonstrated in gasdynamic test facilities for either aerodynamic or aeropropulsion applications. Both offer specific advantages for flows which have limited optical access or are highly luminous and particle laden.

Stimulated Raman Spectroscopy and Its Application

The stimulated Raman scattering process is analogous to spontaneous Raman scattering generated by a pump laser but it includes a second probe laser beam with its frequency shifted so that the difference frequency is resonant with a Raman transition of the molecule. Radiative power is then converted both in frequency and in propagation direction, from the laser beam of higher frequency to the beam of lower frequency, by the stimulated Raman interaction. If the probe laser is a high-power, cw ion laser and the pump laser is a pulsed, high-energy, tunable dye laser, the interaction is observed as a small gain or loss modulation in the probe beam intensity. The modulation intensity is proportional to the product of the intensities in each laser during the pulse duration and the SRS signal is usually taken as the time integral of the modulation component. The SRS signal varies linearly with the density of the scattering species at low densities where the Raman transition is predominately Doppler broadened and it is independent of density when collision broadening is dominant. Finally, the crossing angle between the two beams may be chosen arbitrarily without regard to the phase matching restrictions which are necessary for other coherent interactions.

On the one hand, the use of SRS is attractive because of the linearity of the interaction with each incident laser intensity and its independence of their phase relationship. This behavior simplifies both the optical system required to normalize the SRS measurements to remove fluctuations in laser intensity and the analysis required to determine the gasdynamic parameters being measured. Moreover, because the signal strength is linear with gas density at low densities, the corresponding reduction in signal is no worse than any other spectroscopic measurement technique. That behavior is advantageous for the detection of minor species and for measurements in high-speed wind tunnels where freestream densities can be very low. As a result, SRS has been used very effectively for detailed spectroscopic studies of a wide range of molecules and it is often preferred to spontaneous Raman spectroscopy when high spectral resolution or spectra from minor species is required.

On the other hand, with the optical arrangement just described, SRS signals appear at the detector as a very small ac modulation superimposed on a large dc photocurrent. Simple ac-coupled electronic circuits can be used to isolate the modulation component but the photon-statistical noise contained in the signal is proportional to the square root of the total photoelectron count in the modulation pulse, including its dc component. Thus, the signal-to-noise ratio for SRS signals is degraded significantly by the dc component. As a consequence, the achievement of accurate measurements generally requires reliance on temporal signal averaging.

For aerodynamic applications of SRS, Exton and Hillard[97] and Exton et al.[98] developed a unique optical arrangement that minimized the effects of a high-vibration environment and allowed measurements in a large wind tunnel. They demonstrated time-averaged point measurements of velocity, temperature, and pressure in a 1.2-m-wide supersonic flow for freestream Mach numbers from 2.3 to 4.6. Similar measurements were obtained in the supersonic flowfield over a flat plate at an angle of attack. A pulsed, narrowband, tunable dye laser was used in combination with a cw Ar^+ laser. The tunable laser was slowly scanned to measure the center frequencies and spectral shapes of the nitrogen Q-branch ($J'' = 10$) transition in both the forward and backward directions. Streamwise velocity was determined by comparing the Doppler shifts of the Raman central frequency in both directions, whereas temperature and pressure were deduced from a comparison of the spectral widths.

Coherent Anti-Stokes Raman Spectroscopy (CARS) and Its Applications

In the past decade, the CARS technique has become highly developed and its capabilities are well characterized. Recent reviews of the detailed physics of CARS and its wide range of successful applications for combustion research and high-temperature gas measurements have been provided by Hall and Eckbreth,[99] Greenhalgh,[100] and Eckbreth.[2] A very recent discussion of some of the remaining problems associated with CARS and some possible solutions is given by Attal-Tretout et al.[101] for combustion applications.

At room conditions, CARS signals are typically orders-of-magnitude more intense than those from spontaneous Raman scattering. They emerge from the sample volume in a beamlike manner that allows all of the signal to be collected and to be spatially and spectrally separated from the energetic laser beams. Single-pulse, simultaneous point measurements of temperature, density, and multiple species concentrations have been demonstrated. Typically, temperature measurements can be achieved with an uncertainty of only a few percent whereas the uncertainty in species densities is often 10% or greater. Minor species can be observed down to a concentration that is limited by the ubiquitous presence of a nonresonant CARS background. For mixtures containing H_2, which is one of the most favorable species for Raman detection, concentration measurements below 100 ppm have been achieved. While not limited to heated or high-pressure applications, accurate CARS measurements based on vibrational Raman transitions are obtained most successfully from gasdynamic environments

at pressures greater than 0.1 atm and temperatures greater than 500 K. These conditions are more prevalent in combustion and aeropropulsion flows than in high-speed aerodynamic flows.

CARS is also a particularly complex laser spectroscopic technique both to implement and to analyze. It requires high-energy beams at two or more frequencies which have well-behaved spectral, temporal, and coherent properties. The optical configuration is similar to SRS but both lasers are pulsed and operated at high energy. Whereas one frequency may be chosen for convenience (e.g., frequency-doubled Nd:YAG at 532 nm), the second beam must either be narrowband and shifted in frequency from the first by the frequency of a Raman transition or have a bandwidth that is large enough to encompass the band of Raman frequency shifts of interest. Contrary to SRS, optical phase matching and proper polarization orientations of the laser beams must be provided, which places restrictions on the configuration and crossing angles of beams arriving at the sample volume.

Also contrary to SRS, the CARS signal varies in a complex manner with density. For example, in the limit of low densities where Doppler broadening is dominant, the CARS signals from isolated transitions vary quadratically with density. In the opposite extreme at high densities, where collision broadening is dominant, the CARS signals from isolated transitions approach a linear variation with density. However, the density dependence is also influenced by the coherent interference of overlapping transitions. The contributions to intensity from each transition at a selected frequency do not simply sum as in linear interactions. Their interference is influenced by both the magnitude and phase of each transition, as described by the real and imaginary parts of a complex nonlinear susceptibility, and by a similar interference from the nonresonant CARS background. As a result, Roh and Schreiber[102] have demonstrated that the density dependence of the signal is often not well described by a simple power law variation based on an integer exponent, except for certain species in certain density ranges. At high densities, collisional narrowing processes also affect the CARS spectrum.[100] Thus, the interpretation of the CARS signal to determine species density requires accurate modeling of all of the processes affecting the CARS signal strength.

The optical systems used for CARS also tend to be more complex than those for most other methods. The spectral energies of the CARS signals are proportional to the product of laser pulse energies at three frequencies, two of which may be identical and from the same laser. Thus, in its degenerate configuration, the CARS signal depends quadratically on the pulse energy of the pump laser and linearly on that of the probe laser. This behavior complicates the optical system required to normalize the effects of pulse energy variations from each laser. Furthermore, because the process is directly dependent on the coherence of the interacting beams, the reduction of effects caused by phase variations is also necessary. Fortunately, several practical approaches have been developed[2,99] for achieving all aspects of signal normalization and they have been successfully used in extreme measurement environments.

Despite the complexities associated with the practical application of CARS, the technology has been persistently advanced to the point where all of the apparent major sources of uncertainties have been identified and their effects have been reduced to an acceptable level for most applications.[2] Research efforts leading to further advancements of the CARS technique are now centered mainly on secondary effects which influence the measurement accuracy and spatial resolution.[101] The necessary physical models have been developed and their parameters determined for most species of major importance. Well-documented and validated computer codes are also available which allow CARS spectra to be accurately analyzed for most gasdynamic applications of interest, including high-density and high-temperature environments.[103] As a result of these advances, quantitative measurements can be made in environments where similar measurements using other approaches are impractical. CARS is particularly advantageous for measurements of temperature and multiple species densities in flows that have limited optical access, are highly luminous, or are particle laden. For aeropropulsion applications, measurements in combustors and rocket exhausts are examples for which CARS is most likely to be a superior approach. Its aerodynamic application to clean, high-speed flows at low densities and low temperatures is probably not the most advantageous in most cases.

Applications of CARS are numerous and widely varied. They include measurements in the combustion chamber of internal combustion engines, at the burning surface of solid rocket propellants, and within the glass envelope of a tungsten-filament lamp—to name a few which demonstrate the capabilities of CARS measurements in applications other than with flowing gases.[2]

There is no surprise that nearly all of the applications to flowing gas measurements have occurred in heated, high-density flows with combustion. One of the first examples was reported by Eckbreth[2] and Eckbreth et al.,[104] who demonstrated the use of CARS for measurements in the exhaust of an afterburning jet engine. The initial test provided time-averaged measurements of temperature and species and demonstrated the capability for remote operation of a CARS instrument in an adverse environment. Following the initial demonstrations, the instrument was upgraded to permit instantaneous measurements at a 20-Hz repetition rate. The capability of the technique was indicated by the acquisition of a histogram of temperature, taken with the engine operating at full augmentation. An advanced CARS system, mounted on a mobile cart, was subsequently developed by Anderson et al.[105] It is capable of withstanding high acoustic and mechanical vibration amplitudes and is remotely operable. It was first applied to obtain single-pulse measurements of temperature in a supersonic combustion facility.

More recently, Antcliff et al.[106] have applied CARS to measure temperatures and species densities in the plume of a supersonic, freejet of burning hydrogen in air. Pointwise mapping of the flowfield was done to provide time-averaged data for comparison with numerical codes, including temperature, nitrogen density, and oxygen density. Further advances in the instrumentation and its application are reported by Antcliff et al.[107] and by Taylor et al.[108]

Although CARS appears to be a complex technique that requires some dedicated effort to implement, its optical installation and operation are not significantly more difficult than that required for the commercial LDV systems that are in widespread use. Furthermore, the technology is now well documented and a comprehensive software library is readily available for its analysis.

Closing Remarks

This overview should give the reader a sense of the demonstrated and potential capabilities of laser spectroscopic methods for aerodynamic applications. Further developments can be expected to occur rapidly. Meanwhile, the critical need has arisen to move many of these advanced measurement capabilities from bench-top laboratory demonstrations to the aerodynamic test environments of large, ground-based facilities and to flight applications. The challenges of that objective can often be more difficult than the development of new measurement concepts because of the additional restraints placed on the control and embodiment of the experimental arrangement.

The requirements of aerodynamic measurement techniques for high-speed flight applications are particularly noteworthy. They introduce a host of unique issues that have not yet been addressed widely. Examples of such issues have been recently described by Cattolica et al.,[109] who examined the measurement capabilities that can be achieved using state-of-the-art, electron-beam and LIF technology for the diagnostics of the shock layer and boundary-layer flows surrounding a small

research vehicle in hypersonic flight. Among other things, their work provides an example of a practical approach to such applications. From it, the kind of additional laser development and spectroscopic studies that will be needed to accomplish some of the unique measurement objectives associated with flight experiments are also identified. Similar studies can be expected to have significant influence on the direction and further development of advanced aerodynamic measurement technology.

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