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REVIEW ARTICLE

Diode laser absorption sensors for gas-dynamic and combustion flows

Mark G Allen†

Physical Sciences Inc., 20 New England Business Center, Andover, MA 01810, USA

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Abstract. Recent advances in room-temperature, near-IR and visible diode laser sources for tele-communication, high-speed computer networks, and optical data storage applications are enabling a new generation of gas-dynamic and combustion-flow sensors based on laser absorption spectroscopy. In addition to conventional species concentration and density measurements, spectroscopic techniques for temperature, velocity, pressure and mass flux have been demonstrated in laboratory, industrial and technical flows. Combined with fibreoptic distribution networks and ultrasensitive detection strategies, compact and portable sensors are now appearing for a variety of applications. In many cases, the superior spectroscopic quality of the new laser sources compared with earlier cryogenic, mid-IR devices is allowing increased sensitivity of trace species measurements, high-precision spectroscopy of major gas constituents, and stable, autonomous measurement systems. The purpose of this article is to review recent progress in this field and suggest likely directions for future research and development. The various laser-source technologies are briefly reviewed as they relate to sensor applications. Basic theory for laser absorption measurements of gas-dynamic properties is reviewed and special detection strategies for the weak near-IR and visible absorption spectra are described. Typical sensor configurations are described and compared for various application scenarios, ranging from laboratory research to automated field and airborne packages. Recent applications of gas-dynamic sensors for air flows and fluxes of trace atmospheric species are presented. Applications of gas-dynamic and combustion sensors to research and development of high-speed flows aeropropulsion engines, and combustion emissions monitoring are presented in detail, along with emerging flow control systems based on these new sensors. Finally, technology in nonlinear frequency conversion, UV laser materials, room-temperature mid-IR materials and broadly tunable multisection devices is reviewed to suggest new sensor possibilities.

1. Introduction

The application of diode lasers to sensing of combustion-generated pollutant emissions began shortly after the demonstration of direct current injection semiconductor lasers in the late 1960s. By 1974, portable sensors were being used to detect CO emission from individual automobiles on US highways (Ku *et al* 1975). Shortly thereafter, diode lasers found application to *in-situ* measurements of combustion gases (Hanson *et al* 1977). Applications of mid-IR diode lasers to combustion and high-speed flow research continue to appear in the scientific and engineering literature, including monitors for ammonia in coal combustion (Silver *et al* 1991), NO in high-enthalpy shock tunnel flows (Mohammed *et al* 1996) and aeroengine exhaust analysis (Wiesen *et al* 1996).

† E-mail address: allen@psicorp.com

Today diode lasers dominate the overall laser market, representing over $1.6 billion in 1996 sales (Steele 1997). The majority of these sales are in the telecommunications industry, comprising devices operating between 950 and 1500 nm. The requirements of this industry for devices operating near room-temperature with wavelength tuning, high spectral purity, long-term stability, and compatibility with fibreoptic distribution networks are all consistent with practical spectroscopic absorption sensors, as well. The volume, quality and wavelength range of room-temperature devices continue to grow rapidly. The large market driving this innovation ensures that rapid progress in laser sources will continue for the next few years.

Recently, adaptation of room-temperature diode lasers to measure gas-dynamic properties in reacting and non-reacting flows has received considerable attention. In this review, we describe this progress and attempt to outline the most promising areas for industrial application and further research. Optical sensing of flow phenomena in general grew rapidly during the 1980s and at least some of the current interest in diode laser sensors may be attributed to the success of these efforts. Using broadly tunable, usually high-power pulsed lasers, researchers demonstrated quantitative measurements of species concentration, temperature, velocity, pressure and other gas-dynamic properties, often with high temporal and spatial resolution, in single-point, two-dimensional or three-dimensional geometries (Eckbreth 1981, Hanson 1986, McKenzie 1993, Kohse-Hoinghaus 1994). The impact of this measurement technology on modern research in gas-dynamic and combustion-flow research has been profound. Its utility in research and development environments is now assumed and has spread to government, university and industrial research laboratories worldwide.

The diode laser demonstrations of gas sensing in the middle 1970s did not lead to such immediate growth and impact. In part, this was due to the nature of the devices themselves. The lasers available at that time were generally based on Pb-salts and operated at multiple wavelengths simultaneously (multi-longitudinal mode) in the 5–15 μm wavelength region. A dispersive spectrometer was usually required to isolate a single laser mode. They operated at a few tens of K, requiring cumbersome closed-loop cryogenic coolers, and only generated tens of μW of optical power. Detectors in the mid-IR also required cooling to liquid N_2 temperatures. These operational complexities, coupled with the generally low device reliability and tedious practical issues associated with aligning optical systems with μWs of IR power, cast diode lasers at a considerable disadvantage compared with the newly emerging high-power tunable dye lasers operating in the visible and UV.

The widespread application of high-power laser diagnostic techniques to combustion and gas-dynamic flow research also advanced the general technology of optical measurements. These advances included practical issues such as methods for achieving optical access to often harsh flow environments, considerations of beam distortion due to flow turbulence or shock waves, packaging and maintenance of sensitive optical components in test facility or industrial environments, as well as the emergence of a common vocabulary and mutual understanding between the applied spectroscopist developing new measurement techniques and the engineer responsible for development of the flow or combustion system. Although the progress of measurement technology using large, high-power lasers continues to evolve, it is now generally recognized that the cost, size and operational complexities of these lasers are likely to limit their application to the research laboratory or large-scale aerospace test facility.

The coincidence of this maturation of understanding with the rapidly expanding capabilities in room-temperature, single-mode diode lasers is fortuitous for both technologies. Most of the spectroscopic-based techniques for gas-dynamic measurements of flow properties such as temperature and velocity that were developed with the larger lasers using fluorescence or coherent scattering techniques have analogues in pure absorption. Compact sources the size of a typical 14-pin electronic chip are becoming available with single-mode tuning ranges approaching 100 nm, optical powers of tens mW, stable operating lifetimes of 10^4 h, and costs below $10 000.

As we shall describe in this review, the operational simplicity of these lasers allows a much higher level of automation in the overall sensor design. This is expected to result in even wider penetration of the gas-dynamic and combustion research community since a much lower level of user sophistication is required. Indeed, commercial industrial sensors would appear to be genuinely practical and a number of diode laser-based products are entering the commercial sensor marketplace.

We begin with a brief description of the most common modern diode lasers and describe the impact of the laser material and structure on the operation of a typical sensor. The fundamental photophysics of absorption-based measurements is then reviewed, including extensions to gas-dynamic measurements such as temperature, velocity, and mass flux. The near-IR and visible spectroscopy of important gas-dynamic and combustion species is reviewed and sensitive detection strategies for their measurements described. Typical architectures of diode laser-based sensors are presented, including fibre-coupled and free-space approaches. Aspects of sensor design important for practical applications are described in detail.

Specific examples of sensor applications to air flows and trace atmospheric gas-flux measurements are described. The application of sensors to gas-dynamic, combustion and emissions monitoring is described in detail, focusing on both fundamental measurement capabilities and practical issues associated with large-scale engineering applications. The inclusion of diode laser sensor in combustion and flow control applications is described. The field of diode laser sensors is driven by new technologies in diode laser sources and fibreoptic components. Emerging developments likely to affect sensors are also outlined. Finally, we conclude with a summary of the current state of the art and suggestions for new areas in which further research and development is likely to have a significant impact on the measurement community.

2. Room-temperature diode laser sources

Near room-temperature diode lasers (operating at a temperature accessible with thermo-electric coolers, generally ~250–350 K) are presently available over a range of wavelengths extending from about 630 nm in the visible to about 2.2 μm in the mid-IR. The available ranges are summarized in figure 1, along with the diode laser materials and typical commercial applications. The shortest wavelength InGaAlP lasers produce a beam easily visible to the eye and are used for He–Ne replacements in laser pointers and barcode scanners. AlGaAs lasers in the 780–900 nm wavelength region are the largest volume/lowest cost lasers used for CD data storage and laser printers. A typical laser in these wavelength regions is a virtual commodity product selling for $5–50 per unit, depending on the volume.

The longest wavelength InGaAs lasers grown on GaAs substrates are primarily high-power pump sources for erbium-doped fibre amplifiers (EDFAs) used for repeaters in long-haul telecommunications applications. The largest

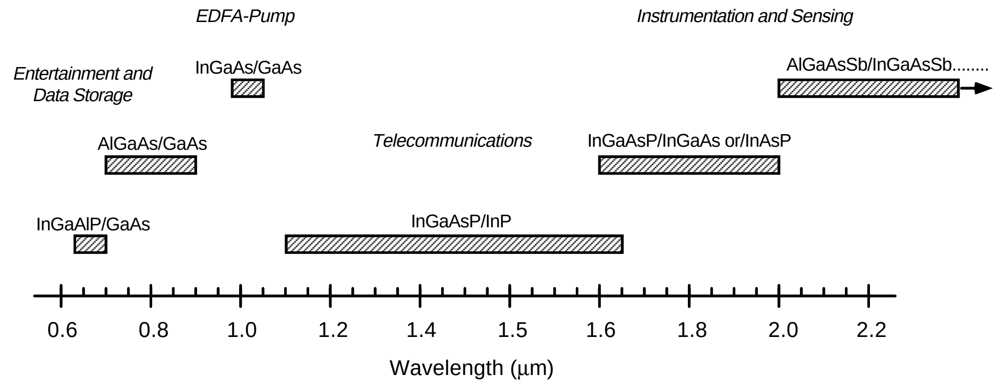

Figure 1. Approximate spectral windows of commercially available, room-temperature diode laser sources.

dollar volume of lasers consists of the InGaAsP devices used for telecommunication transmission. Using InP substrates, devices can be grown with lasing wavelengths between about 1100 and 1650 nm, although industry standards restrict most devices to two spectral windows. The minimum material *dispersion* of silica fibreoptic waveguides at 1.31 μm generates one of these windows and, with improved manufacturing tolerance, most large volume vendors have now reduced the window of scatter in available devices to less than 10 nm about this centre. The minimum material *attenuation* of silica fibreoptic waveguides at 1.55 μm generates the second of these windows. However, this window is considerably broader due to emerging wavelength-division-multiplexing (WDM) standards and the fact that devices are widely available with selectable room-temperature wavelengths over nearly 100 nm about this centre. Prices for fibre-coupled telecommunications lasers range from ~\$2000 for standard wavelengths to ~\$10 000 for custom wavelengths. Output powers from 3 to 10 mW are common.

Sales of diode lasers beyond 1.6 μm represent a negligible component of the overall market, but are important for sensor and instrumentation application, as will be described. Using InGaAs or InAsP substrates allows InGaAsP devices to operate at wavelengths as long as 2.1 μm. Beyond 2 μm, commercial devices are beginning to appear based on antimonide gain and substrate stoichiometries at wavelengths as long as 2.3 μm. Continuous-wave (cw) operation generally requires cooling to about 250 K. Price, performance, and lifetime of these devices vary considerably due to the immature fabrication processes involved.

The basic diode laser structure consists of a short ($\leq$500 μm), cleaved section of laser material with ohmic contacts on the top and bottom. Current injected through the gain medium creates optical gain along the laser axis and the finite reflectance of the cleaved crystal planes is sufficient to create a Fabry–Perot (FP) resonator. These FP structures are common in the visible lasers where the gain medium tends to oscillate in a single-longitudinal mode defined by one order of the resonator. This lasing condition is not necessarily repeatable, however, so that the laser may operate on a different longitudinal mode (and hence laser wavelength) from one day to the next at the same operating temperature and current.

All diode lasers can be wavelength tuned in two ways. By adjusting the temperature of the diode, the effective optical index of the waveguide is changed, thereby adjusting the resonant condition of the laser cavity. Using this approach, each laser can typically be tuned from 3 to 5 nm between 275 and 325 K. For FP lasers, this tuning range is typically not continuous and exhibits hysteresis and regions of multimode instability (Franzke *et al* 1993). Continuous tuning range between mode hops may be as small as 0.1 nm. Because of the thermal mass of the diode and heater/cooler element, this type of tuning is usually restricted to a few Hz maximum bandwidth. The second mode of wavelength tuning involves the injection current density into the gain section. At any given temperature, the injection current can be varied to modulate the optical index. Of course the output power is also modulated, but the laser wavelength may be tuned extremely rapidly. Injection current modulation bandwidths on the order of 10 GHz are achieved in telecommunications lasers. Typical FP InGaAlP and AlGaAs lasers tune at a rate of about 0.15 Å mA^{-1} or about 0.25 cm^{-1} mA^{-1} near 760 nm. The total continuous tuning range is highly device dependent and a function of the laser gain medium and fabrication technique. For visible FP lasers, 1–2 cm^{-1} is common with 5–10 cm^{-1} of inaccessible wavelengths between mode-hops.

The discontinuous tuning range and lack of reproducibility of the operating conditions of FP lasers are unattractive for many sensing applications. In some cases, a wavemeter may be required to determine the laser operating wavelength at the nominal set-up temperature and injection current. The most common method for improving the wavelength control of diode lasers is to form a spatially periodic modulation of the refractive index in the material adjacent to the gain medium, enhancing the cavity feedback at a particular wavelength through evanescent coupling of the lasing mode. These distributed feedback (DFB) lasers have been grown at wavelengths as short as 760 nm (Morris *et al* 1995) and are commonly available in InGaAsP devices out to 2 μm. They can be temperature tuned over 3–5 nm about the nominal operating wavelength although the injection current tuning rate is typically below 0.1 Å mA^{-1}.

The injection current tuning range is 1–2 cm^{-1} and limited by lasing threshold at the low end and output facet damage at the high end. The long interaction lengths of the DFB grating provide very narrow linewidths, however, generally on the order of 10 MHz in single-section devices and as low as a few hundred kHz in multisection devices (see section 6).

The wavelength-selective and gain sections of the laser may be separated, thereby decoupling the output power from the wavelength tuning. Such devices are termed distributed Bragg reflector (DBR) lasers. Here, the periodic index modulation is applied to sections at the rear (usually) of the laser and electrically isolated from the gain medium. High-power, fixed-wavelength lasers used for optical pumping are typically DBR structures, although multi-electrode lasers with separate gain and wavelength control are beginning to be applied to gas sensing (Larson *et al* 1997, Upschulte *et al* 1998) and are expected to have a broad impact on diode laser sensors. Because the wavelength tuning range is no longer restricted by the threshold/damage limits of the gain medium, wide current tuning ranges are possible (Yamaguchi *et al* 1985).

An alternative approach for single-mode wavelength control is to build an external resonator around the diode laser, treating the diode as simply a current-pumped gain medium—analogous to an optically pumped dye cell in a conventional dye laser. Uncontrolled optical feedback into diode lasers is typically very deleterious to the diode laser's frequency and amplitude stability (Goldberg *et al* 1982, Cassidy and Bonnell 1988). Reflections from optical elements such as lenses, prisms or cleaved fibre facets as low as −40 dB can induce severe power instabilities, multimode lasing, or linewidth broadening. Controlled feedback, however, can lock the laser wavelength to the external cavity mode. Using an adjustable, wavelength-selective element such as a grating, the laser wavelength may be tuned over most of the gain medium. Operational details of the external cavity diode laser (ECDL) are sensitive to the construction of the cavity and the reflectance properties of the surfaces within it (Ventrudo and Cassidy 1990). Commercially available ECDLs (Nguyen *et al* 1994, Oh and Hovde 1995, Sonnenfroh and Allen 1997a) generally construct a Littman cavity between the rear facet of the diode, a tunable grating, and a high-reflectivity mirror. Such ECDLs are available with centre wavelengths from 760 to 1550 nm and tuning ranges from 30 to nearly 100 nm. The tuning performance of the laser is critically dependent on the quality of the antireflective coating on the front facet of the diode, however, since weak reflectance from this facet can set up a second set of cavity modes leading to mode-hops in the tuning range or coupled frequency, polarization and amplitude modulation of the output with tuning (Sonnenfroh and Allen 1997a). Tuning rates are limited by the inertia of the tuning elements to a few kHz and output powers are generally a few mW. Since the ECDLs are physically much larger than simple current- and temperature-tuned devices and require mechanical motion to operate, their suitability for long-term, remote sensor applications is unclear. Their broad tuning range and relatively high output power, however, result in extremely useful laboratory tools.

3. Absorption spectroscopy using room-temperature diode lasers

3.1. Absorption fundamentals

Diode laser sensors are based on absorption of the wavelength-tuned laser intensity as the beam propagates across the measurement path. In contrast to some laser-induced fluorescence, coherent scattering, or other techniques commonly applied to combustion and flow diagnostics, the measurement is easily quantified. The absorption is described by the Beer–Lambert relation:

$$I_\nu = I_{\nu,0} \exp[-S(T) g(\nu - \nu_0) N \ell] \qquad (1)$$

where I_ν is the monochromatic laser intensity at frequency ν, measured after propagating a pathlength ℓ through a medium with an absorbing species number density N. The strength of the absorption is determined by the temperature-dependent linestrength, $S(T)$, and the lineshape function, $g(\nu - \nu_0)$ (Arroyo and Hanson 1993). The lineshape function describes the temperature- and pressure-dependent broadening mechanisms of the fundamental linestrength. The product of the linestrength and lineshape function is the absorption cross section. The temperature dependence of the linestrength arises from the Boltzmann population statistics governing the internal energy-level population distribution of the absorbing species. If the medium is invariant over the absorption pathlength, then straightforward inversion of equation (1) yields a quantity proportional to a number of gas-dynamic properties.

The linestrength of the absorption transition is a fundamental spectroscopic property of the absorbing species (cf Baer *et al* 1996), although it is usually expressed in relation to tabulated values available in one of a number of databases. The most commonly used and extensive database for IR transitions of small molecules is the US Air Force HITRAN database (Rothman *et al* 1992), originally developed for atmospheric transmission applications. The linestrength at any temperature $S(T)$ can be calculated from the known linestrength at temperature $S(T_0)$ using

$$S(T) = S(T_0) \frac{Q(T_0)}{Q(T)} \exp\left[-\frac{hcE}{k}\left(\frac{1}{T} - \frac{1}{T_0}\right)\right] \times \left[\frac{1 - \exp(-hcE/kT)}{1 - \exp(-hcE/kT_0)}\right] \qquad (2)$$

where Q is the total molecular internal partition function, E is the energy of the lower transition state, h is Planck's constant, k is Boltzmann's constant and c is the speed of light (Rothman *et al* 1992). The last term accounts for stimulated emission and is negligible at wavelengths below 2.5 μm and temperatures below 2500 K.

The lineshape function describes the effect of collisional and thermal processes on broadening the molecular transition frequency. Detailed discussions of lineshapes relevant to near-IR and visible laser absorption can be found in the literature (Arroyo and Hanson 1993, Upschulte and Allen 1997). For *in-situ* measurements near atmospheric pressure, the lineshape function is usually described by a Voigt profile containing both collisional and thermal broadening. Typical values of the full width at half maximum (FWHM) of lineshapes relevant to gas-dynamic and combustion-flow sensing are generally on the order of 0.15 cm^{-1} (4.5 GHz) at atmospheric pressure. Hence, diode lasers with spectral linewidths below 10 MHz can generally be considered monochromatic with respect to the absorption lineshape.

If the gas temperature, line-strength and absorption path are known, the measured transmission may be directly related to the absorbing species number density. It is usually possible to select a particular absorption transition such that the temperature variation of the linestrength over some limited range (typically several hundred K) can be neglected. Separate measurements of temperature can be used to correct for variations, if necessary.

Alternatively, two absorption transitions may be probed (using one or two lasers, depending on the target transition separation and the laser tuning range). The ratio of the integrated absorbance of each transition is a pure function of temperature:

$$R = \left(\frac{S_1}{S_2}\right)_{T_0} \exp\left[-\frac{hc\Delta E}{k}\left(\frac{1}{T} - \frac{1}{T_0}\right)\right] \tag{3}$$

where S_1 and S_2 are the linestrength values at some reference temperature, T_0, and ΔE is the energy separation of the absorbing states. The temperature sensitivity depends on the values of the reference linestrengths and the energy separation. With the temperature determined, either or both absorbances can be used to determine the number density (Arroyo and Hanson 1993, Allen and Kessler 1996, Baer *et al* 1996).

If the laser beam propagates across a flow with a bulk velocity V, the molecules in the moving reference frame of the gas observe a laser frequency that is Doppler-shifted according to

$$\Delta\nu_{\mathrm{Doppler}} = \frac{V}{c}\nu_0 \cos\theta \tag{4}$$

where θ is the angle between the laser propagation and bulk flow directions. For near-IR diode lasers, the magnitude of the Doppler-shift is approximately 3×10^{-5} cm^{-1} per m s^{-1} of flow velocity. At typical atmospheric pressure conditions, a flow velocity of 1 m s^{-1} represents a shift-to-width ratio of about 10^{-4}. Since the absorption measurement itself gives the density of the gas, measuring the Doppler-shift allows determination of the density–velocity product or the mass flux (Philippe and Hanson 1993, Arroyo *et al* 1994a, Miller *et al* 1996).

3.2. Overview of near-IR and visible spectroscopy of gas-dynamic and combustion species

Absorption-based sensors have the highest sensitivity and selectivity when a spectrally narrow source is used to probe a spectrally narrow feature. Tuning the wavelength of the source across the absorption feature distinguishes the isolated feature from background absorption, scattering, or extinction effects due to obscuration of the optical path or changes in the total source power coupled onto the receiver. Thus, most applications relevant to gas-dynamic and combustion flows are based on absorption by low-molecular-weight molecules with well resolved absorption transitions such as O_2, H_2O, CO, CO_2, NO, NO_2, OH, NH_3, HF, H_2S, and CH_4. With the exception of visible transitions of O_2 and NO_2, the absorption measurements are performed on overtone and combination vibrational absorption bands. Typical linestrengths of these transitions are between 10^{-23} and 10^{-21} cm/molecule, two to three orders of magnitude below the fundamental vibrational transitions in the mid-IR.

Water vapour is a trace species in atmospheric air and a major product species of all hydrocarbon combustion. It possesses $\nu_1+\nu_3$, $2\nu_1$, and $2\nu_3$ absorption bands throughout the 1.3–1.4 μm region, including well-resolved lines at the technically important wavelengths near 1.31 μm (Allen and Kessler 1996). Custom-wavelength lasers are available at 1.39 μm which access transitions approximately 300 times stronger at room temperature (Allen *et al* 1995). Other bands are ubiquitous throughout wavelengths between 1 and 2 μm and can be particularly important as interferences in combustion gases at high temperature, where transitions from higher-lying rotational and vibrational energy levels become strong. For example, strong $\nu_1 + \nu_2$ and $\nu_2 + \nu_3$ transitions bands are also available between 1.8 and 1.9 μm (Sonnenfroh and Allen 1997b). As is typical of the relatively weak absorption transitions in the near-IR, little is known about the line positions, assignments, and strengths of some of these transitions. A recently released high-temperature version of HITRAN, the HITEMP database (Rothman *et al* 1997), appears to address some of the shortcomings of the water database, but has yet to be extensively confirmed with experimental measurements except in the region near 1.31 μm and is only valid to 1000 K (Upschulte and Allen 1997).

A number of molecules of interest in combustion flows possess transitions near the important 1.55 μm spectral window. Carbon monoxide has a second overtone band centred at 1.575 μm (Cassidy and Bonnell 1988, Hanson 1997, Sonnenfroh and Allen 1997a, Mihalcea *et al* 1997, Gabrysch M *et al* 1997). The $3\nu_1 + \nu_3$ band of CO_2 is centred near the CO second overtone at 1.575 μm and the $2\nu_1 + \nu_3$ band is centred at 2.01 μm with transitions two orders of magnitude stronger (Hanson 1997, Sonnenfroh and Allen 1997a). Recent measurements of the high-temperature CO_2 bands near 1.57 and 2.01 μm suggest that the HITEMP database may substantially over-predict the absorption strengths at flame temperatures (Kessler *et al* 1997). The OH radical's first overtone band extends throughout this region with linestrengths at typical flame conditions of $\sim 5 \times 10^{-21}$ cm/molecule and is easily detected in laboratory flames (Sonnenfroh and Allen 1996a, Upschulte *et al* 1998). The methane $\nu_2 + 2\nu_3$ combination band near 1.3 μm has been used for absorption sensors (Silveira and Grasdepot 1996), as well as the stronger $2\nu_3$ band near 1.65 μm (Shimose *et al* 1991, Uehara and Tai 1992, Hovde *et al* 1995, Nagali *et al* 1996a, Chou *et al* 1997, Mihalcea *et al* 1997). Second overtone transitions of NO near 1.8 μm have recently been identified and detected in ambient and combustion gases using a DFB laser (Sonnenfroh and Allen 1997b).

Water vapour, acetylene and other molecules possess weak, higher-order overtones in the visible spectrum and have been detected using AlGaAs lasers, although near-IR sensors achieve a lower detection limit. Elsewhere in the red portion of the visible, electronic transitions of some molecules have been used for diode laser sensors. Molecular oxygen possess a spin-forbidden electronic system whose $(0,0)$ band near 763 nm has proved important for many gas-dynamic and combustion applications (Bruce and Cassidy 1990, Philippe and Hanson 1993, Nguyen *et al* 1994, Miller *et al* 1996). A minor constituent of combustion-generated NO_x, NO_2, has a broad and poorly understood electronic system throughout the visible spectrum. The weak red end of this spectrum is accessible with ~635 and ~670 nm lasers, both of which have been used to detect trace quantities of the gas (Mihalcea *et al* 1996, Sonnenfroh and Allen 1996b).

Many other molecules of possible interest in combustion or gas-dynamic measurements have been detected using visible and near-IR diode lasers. The recent review by Feher and Martin (1995) provides a good overview of the spectroscopy of these species.

3.3. High-sensitivity absorption measurements

Because of the weak absorption strengths of the near-IR and visible features described above, most diode laser sensors must have extremely high sensitivity to small changes in the transmitted power—typically less than 1% attenuation for major species like O_2 and H_2O and on the order of parts in 10 000 for minor species such as CO and NO. Direct absorption, even using two detectors to compare common changes in the incident and transmitted intensity is generally limited to absorptions greater than 1%. The oldest and most common approach for sensitive absorption measurements with diode lasers involves high-speed modulation of the laser injection current, introducing amplitude and frequency modulation on the output beam. These techniques are broadly termed frequency-modulating (FM) spectroscopy. Using phase-sensitive detection at some harmonic of the modulation frequency, background-equivalent absorbances of $\sim 10^{-7}$ have been demonstrated (Bjorklund 1980, Lenth 1983, Carlisle and Cooper 1989).

If the average wavelength of the modulated laser is fixed at the centre of the absorption lineshape, the peak FM signal contains contributions from laser modulation parameters as well as pressure- and temperature-dependent line-broadening mechanisms. Scanning the diode laser wavelength across a fully resolved lineshape and integrating the resulting absorption signal can be used to remove the pressure and temperature contributions to the lineshape, although the integrated absorbance may still be a function of laser parameters such as modulation depth and frequency. Such scanning is usually desirable in any case, since it establishes a reliable baseline which may be varying across the laser scan range. The total range over which the centre frequency of the modulated signal may be continuously tuned is highly device dependent. This range is reduced in the presence of frequency modulation. For this reason, many ultrasensitive FM measurements are made in temperature- and pressure-controlled cells which operate below atmospheric pressure, although field sensors based on FM spectroscopy have been deployed by several groups for ground-based monitors at atmospheric pressure (cf Silver and Hovde 1994).

In principle, dual-beam approaches which divide the signal beam (that which passes through the absorbing medium) by a reference beam (which does not pass through the absorbing medium) should allow cancellation of common-mode excess laser amplitude noise. In practice, however, conventional dual-beam measurements are severely limited by the balancing requirements in the two detection channels. Generally speaking, the common-mode noise on the laser source will be transmitted through the balancing circuit at a level proportional to the product of the noise-to-power ratio and the percent of imbalance (Houser and Garmire 1994). This will effectively determine the minimum resolvable absorption level. For example, a laser source with a 1% amplitude noise in the target detection bandwidth must have signal and reference channels balanced to 10^{-4} to achieve a noise level, and minimum detectable absorption, of 10^{-6}. Even if this balance could be achieved in a laboratory set-up, it would be impractical to maintain in a field instrument. Furthermore, spatial variations in detector responsivity, drifts in amplifier or other circuit characteristics, changing intensities in the two beams due to polarization or reflectance variations during a laser scan, and lack of high-bandwidth temporal coherence in transimpedance amplifiers all contribute to additional noise on the voltage ratio.

An alternative approach invented by Hobbs (1997) replaces this optical balancing limitation with an electronic photocurrent balancing. This approach has been adapted for a variety of diode laser sensors (Allen *et al* 1995, Allen and Kessler 1996, Miller *et al* 1996, Sonnenfroh and Allen 1996b) and has demonstrated equivalent sensitivity to FM-based schemes, with over 50 dB of common-mode noise cancellation. It has advantages in some applications in that the pure absorption lineshape is recovered and no high-frequency electronics are required. With either FM or balanced-detection approaches, the practical sensitivity limit is usually determined by optical interferences or weak etalon effects from optical elements or fibre components in the sensor configuration.

4. Sensor architecture

A typical fibre-coupled, multiwavelength diode laser sensor configuration is shown in figure 2. The temperature of each laser is controlled using thermo-electrically stabilized mounts and commercially available closed-loop controllers. The injection current into each laser is also controlled using commercially available, highly stable current sources. These units can be purchased in either benchtop or single-card versions suitable for integration in standard computer or electronic racks. The most convenient packaging for the laser itself is the communications-style fibre pigtail package wherein the laser chip, Faraday isolator, antireflection coated lens, and fibre are all pre-aligned and soldered into a common package about the size of typical 14-pin electronic chip. The entire assembly is mounted on a common thermo-electrically cooled mount. These packages are routinely available for InGaAsP lasers and can be assembled on a custom basis for some other wavelengths.

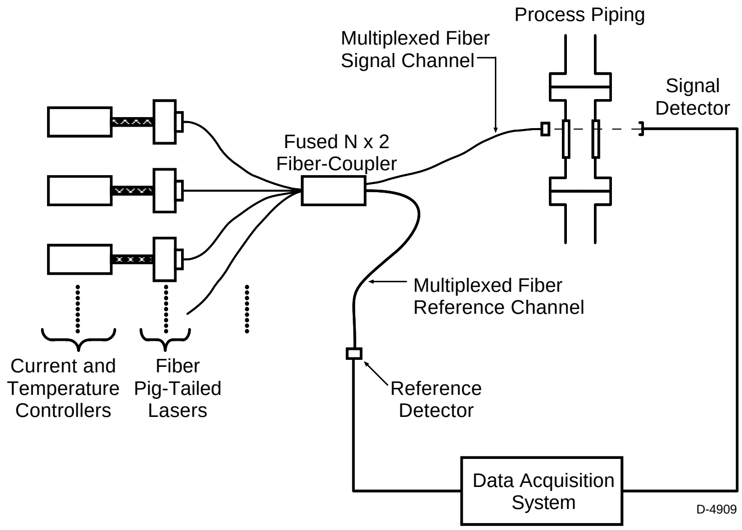

Figure 2. Schematic of typical multiwavelength diode laser sensor configuration.

The details of the laser packaging can be critically important for many applications. Fibre-coupled systems are often preferred for their simplicity and the ease with which the laser light can be transported over distances of hundreds of metres. For stable transmission, single-mode fibres, where a single transverse mode structure is supported in the waveguide, are superior. Larger diameter, multimode fibres tend to exhibit uncontrolled bending losses and interference effects associated with multiple transverse modes. Depending on the guided wavelength, the core diameter for single mode fibres ranges between 5 and 10 μm, introducing extreme alignment tolerances at the coupler. Careful attention by the manufacturer to alignment and thermal stabilization of this alignment is critical for a well-behaved device.

An optical isolator is also critical to good performance. Because of the laser's high sensitivity to small levels of optical feedback and the efficiency with which the fibres transmit back reflections, some method of rejection of the backward propagating light is necessary to maintain stable laser performance. Optical isolators are based on cross-polarizers with a Faraday rotator between. The Faraday rotator uses a fixed magnetic field to rotate the polarized transmission of the first polarizer into the plane of transmission of the second polarizer for light propagating out of the diode laser. Backward propagating light is rotated by the same amount and in the same direction as the forward propagating radiation so that it is not transmitted by the polarizer nearest to the diode itself. A combination of isolators and angle-cleaved fibre end faces with at least -45 dB suppression are preferred. These are generally available for wavelengths between 760 nm and 1.8 μm, but less commonly for shorter or longer wavelength sources at the present time. We find that angle-cut, physical contact (APC-type) connectors give the best results for other fibre–fibre connections in the remainder of the system.

With short or long wavelength devices where isolators or fibre pigtails are not available, the astigmatic, non-circular and highly diverging output of the diode laser must be collected, circularized and collimated (to the extent that the device astigmatism allows) using specially designed aspheric lenses. Antireflection coatings of these lenses is critically important due to the lack of an isolator.

The configuration shown in figure 2 is for a balanced detection set-up where signal and reference channels are required. Taking advantage of fibre components for wavelength division multiplexing, multiple lasers may be combined onto common signal and reference fibres using fused single-mode $N \times 2$ couplers where N represents the number of laser sources. These devices are all-solid-state components where the cores of each fibre are fused together over a physical dimension sufficient to couple the guided modes onto the output fibres in fixed proportion. Single wavelength, 1×2 versions of these couplers at wavelengths between 760 nm and 1.55 μm have been used in a number of sensor configurations with no loss of sensitivity. Two and three wavelength sources have also been demonstrated using 2×2 and 3×2 couplers (Allen and Kessler 1996, Baer *et al* 1996, Allen *et al* 1997).

The fibre reference channel carries the unattenuated light directly to a reference photodetector, which can be housed with the laser sources. The fibre signal channel transmits the light to be used for the absorption measurement directly to the measurement location. This has numerous obvious advantages for practical sensors, including the ability to remotely locate the laser and control electronics hundreds of metres from the measurement location, to use $1 \times M$ couplers to simultaneous address M measurement locations with a single source, or to rapidly change measurement locations or source lasers without extensive re-alignment of the optical set-up.

Light from the fibre is launched across the detection pathlength using a specially designed lens assembly. Typically, these small lenses (diameter $\sim$0.5 cm) are mounted in a housing directly onto the end of the fibre cable by the manufacturer. The launch lens design parameters are determined by the distance over which a nearly collimated beam is desired, and can include single-pass distances of several metres while maintaining a beam diameter on the order of a few mm. Figure 2 shows a typical exhaust-gas emissions monitor set-up, where the signal channel is launched across a duct in the exhaust-gas stream. Appropriate windows are required, whose material has been chosen to provide good transmission properties at the wavelength of interest. For high-sensitivity applications, antireflection coating of the window at the laser wavelength reduces etalon background fringes in the transmission data. Fused-silica windows are satisfactory for most visible and near-IR applications of moderate temperature. Special IR-grade quartz should be used for applications near 1.4 μm due to a material absorption band in normal quartz that can affect the properties of antireflection coatings and lead to severe etalons.

The transmitted light may be recollected using a capture lens/fibre assembly—essentially the same as the launch optic except operated in reverse. As mentioned earlier, single-mode fibres provide the most stable transmission and immunity from fibre vibration and positional changes, but the severe alignment tolerances required to capture all (or a fixed fraction) of the transmitted light in a $\sim$10 μm core make this a very difficult approach in practical applications. Flow turbulence or facility vibrations can introduce pointing-instabilities in the launch optic which couple to amplitude instabilities in the transmitted radiation and can severely degrade the sensor response. Multimode fibres with 50 or 100 μm core diameters alleviate the pointing-instability problem considerably but introduce other undesirable interference or vibration effects. We find that the most reliable and reproducible response is achieved by directly illuminating the signal photodetector.

Large area Si and InGaAs detectors ($\sim$1–5 mm diameter) are available which reduce the pointing instabilities and maximize the collected signal. In single-ended FM approaches, these alignment tolerances are reduced because the absorption signal is detected in a narrow (temporal) frequency band. Both Si and InGaAs detectors are mature technologies with numerous commercial vendors. Si responsivity is better than 0.5 A W^{-1} from 700 to 1000 nm and InGaAs is on the order of 1 A W^{-1} from 1.2 to 1.6 μm. Extended-red response InGaAs devices are becoming available with good response to 2.1 μm. Compared with Si, however, InGaAs exhibits high dark currents, typically in the nano-amp range compared with subfemto-amps for comparable area Si. Since the noise floor of most sensors arises from laser noise or optical interference effects, this is rarely an issue, although for very low-power sources (<1 μW) it should be considered.

The remainder of the sensor consists of conventional data-acquisition and control electronics. In typical operation, the laser temperature is adjusted to tune the output wavelength to coincide with the absorption transition of interest. The laser injection current is swept across $\sim$10–20 mA using a ramp generator, creating a 1–2 cm^{-1} frequency sweep. This sweep can be accomplished with bandwidths of hundreds of kHz, although typically a few kHz is used to since the overall range of the laser wavelength scan for a given amplitude current ramp is reduced with high-frequency modulation (Philippe and Hanson 1993). For the multiplexed configuration shown in figure 2, each laser is swept at different phases of common ramp function. This provides a time-domain multiplexing where a single photodetector observes the sequential absorption features as each laser is swept across its corresponding lineshape.

Figure 3 is an example absorption scan from a three-wavelength version of the multiplexed configuration shown in figure 2 for simultaneous detection of CH_4, CO_2, and H_2O. The absorption measurements were made in a single pass through a 50 cm cell filled with 0.5 Torr CH_4, 68.1 Torr CO_2, and 14.1 Torr H_2O. The first peak at the left of the figure is a group of three unresolved CH_4 transitions of the $2\nu_3$ band near 1.65 μm. The second peak is an isolated CO_2 transition of the $3\nu_1+\nu_3$ band near 1.57 μm. The last strong peak at the right, near data index 8.5, is a water vapour transition near 1.392 μm and the weak feature at the far right is a neighbouring water transition. The narrow feature near data index 7 is a repeat of the strong water transition recorded during the laser flyback at the beginning of the scan ramp. The mapping between data index and frequency is different between each of the three lasers according to the scan rate (MHz mA^{-1}) differences in each laser, so the relative widths are not representative. The entire scan was recorded using sweeps of 10 ms duration and 200 sweeps were averaged to reduced the bakcground noise, for a total measurement time of 2 s.

As an alternative to time-domain multiplexing, wavelength-domain multiplexing has also been used in diode laser combustion sensors (Baer *et al* 1996). Here, the lasers can be ramped across their respective absorption lines truly simultaneously. Multiple wavelengths in the transmitted beam are separated onto multiple detectors using a frequency-dispersive device such as a conventional reflection grating. Grating demultiplexers may also be useful for isolating the laser wavelengths in high-temperature applications where the measurement gas is emitting at wavelengths that would otherwise be viewed by an unfiltered detector. Wavelength division multiplexing components are beginning to appear commercially for telecommunications applications which combine fibre couplers with filters to separate multiple laser frequencies into individual channels. Compared with gratings, these devices will be much more compact and will require no alignment, but will have a low throughput. Since all of the transmitted light is split into each of the M output channels, but only one channel will be transmitted, the total signal power on any channel is reduced by $1/M$. These devices have yet to appear in a diode laser sensor configuration; therefore any interference or etalon effects they may introduce in the signal are unknown.

Although absorbance sensitivities of 10^{-5} can be achieved in practical installations, for high-sensitivity trace-gas monitoring or applications requiring a high signal-to-noise ratio, it is often convenient to increase the absorption pathlength across the usually fixed physical dimension of the flow facilitiy. The conventional approach for this is to launch the beam into a multipass cell embedded in the flow or measurment environment. Known as Herriot cells

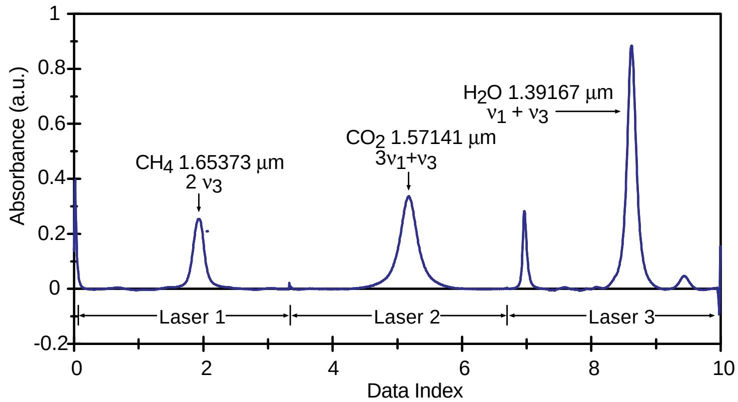

Figure 3. Example near-simultaneous detection of CH_4, CO_2, and H_2O using a three-wavelength, time-domain multiplexed, diode laser sensor operating at 1.65, 1.57, and 1.39 μm.

(Herriot *et al* 1964, Herriot and Schulte 1965), these have been widely applied in atmospheric monitoring and are commercially available with astigmatic optics providing over 100 m of non-overlapping optical pathlength in a physical cell less than 1 m in length. Simplified versions have recently been applied in hypersonic shock tunnel flows (Hanson 1997).

As with all optical diagnostics, optical access to the measurement location is often the most severe engineering challenge for a given sensor. Diode laser sensors suffer the usual constraints, requiring that the windows and window mounts sustain the pressure and temperature of the flow gases. For *in-situ* combustion measurements, film-cooling of the interior window surface with a purge gas is often used to both cool the window and to keep soot or other particulate from impacting or depositing on the surface. In supersonic or hypersonic flows, care must be taken to ensure that shock waves formed from the windows or mounting blocks do not excessively heat the windows, introduce flow-perturbations, or induce severe beam steering effects.

5. Example diode laser sensor applications

5.1. Air flows

The availability of room-temperature diode lasers near 760 nm provides direct access for spectroscopic measurements of O_2 and, therefore, unseeded air flows. Numerous applications for monitoring air-flow properties in wind tunnel and air-breathing combustion systems have been demonstrated. Also, despite the fact that diode laser absorption sensors necessarily provide line-of-sight averages, tomographic reconstruction techniques are well established for inverting multiple lines of sight into spatially resolved measurements (Chung *et al* 1995, Varghese *et al* 1997). Tomographic reconstruction using FM detection of O_2 jets has been demonstrated with density reconstruction uncertainties on the order of 5% over the domain (Kauranen *et al* 1994). Extensions of this technique for simultaneous measurements of air density and temperature using a multiplexed two-laser system has also been demonstrated (Kessler *et al* 1995). Figure 4 is an example reconstruction from this work of the temperature field above a non-axisymmetric jet mixing flow consisting of a 150 K jet of pure N_2 located at $x = 5$ cm and $y = 0$ cm in the domain and mixing with a room-temperature jet of pure O_2. The reconstructed temperature field was compared with detailed thermocouple surveys and exhibited an rms uncertainty of 14 K over the domain. Temperature uncertainty over single, uniform lines of sight was on the order of 3 K. This type of sensor has applications to measurements of temperature and density distortions at aeroengine inlets in large-scale test facilities.

Simultaneous air density and velocity measurements have been significantly refined and applied to a full-scale F-100 aeroengine inlet in ground tests using balanced dual beam detection (Miller *et al* 1996). A single FP AlGaAs laser and 1×4 splitter deployed two signal channels across the 0.92 m diameter inlet, as illustrated in figure 5. 60 m fibreoptic cables delivered the light from an equipment bunker near the test stand and the launched beams directly illuminated large-area Si photodetectors mounted on the opposite side of the inlet. Figure 6 shows single sweep and average O_2 lineshapes recorded with the engine operating a full power (no afterburner) and clearly illustrates the Doppler shift between the two measurement paths. At this power level, the duct air density was 0.91 kg m^{-3} and the flow velocity was 160 m s^{-1}. A large set of data acquired over the full range of engine operating conditions was statistically analysed and demonstrated density, velocity and mass-flux precision on the order of 2% or less and a precision-limited velocity of 40 cm s^{-1} at atmospheric pressure. This corresponds to a Doppler shift/linewidth ratio of 3×10^{-4}. Such high-precision, direct measurements of inlet air flux is required for improved aeroengine control and flight versions are under development.

Diode lasers have been extensively applied to measurements of trace gases in the atmosphere (see the recent review by Feher and Martin 1995). In conjunction with a multipass cell to achieve extreme sensitivity in a relatively short path (∼50 cm) and a conventional directional anemometer to record wind speed and direction, it is possible to make essentially point measurements of the turbulent exchange of trace gases from anthropogenic

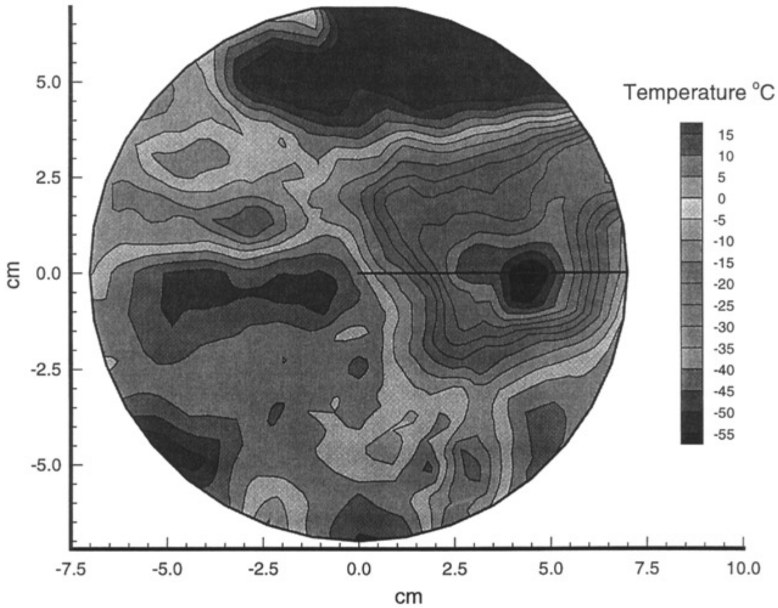

Figure 4. Example tomographic reconstruction of average temperature distribution in a turbulent, mixing O_2/N_2 jet flow.

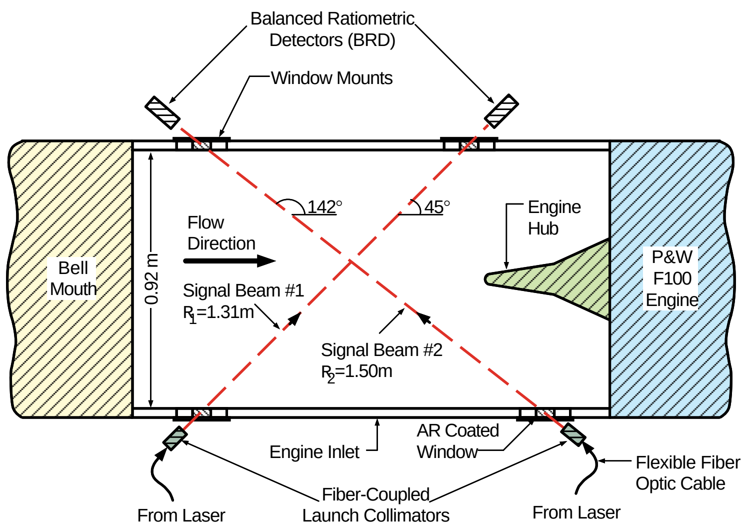

Figure 5. Diode laser air mass flux sensor configuration on full-scale aeroengine inlet.

sources such as waste dumps, agricultural fields, industrial emission, etc within the atmosphere. Using a 1.65 μm DFB laser diode, researchers have recently determined the flux of methane from a landfill with a sensitivity below 100 ppb (Hovde *et al* 1995). Flight versions of 1.39 μm diode laser sensors for trace water vapour measurements have also been demonstrated on research aircraft with sensitivity on the order of 10 ppm (Silver *et al* 1994). Here, the pointlike nature of the measurement is due to the vast scale difference between the ~1 m path and the characteristic dimension of the atmosphere. Using water-cooled, fibre-coupled probes, similar short-path, quasipointwise absorption measurements have been demonstrated with UV absorption in flames using a frequency-doubled ring-dye laser (Kimball-Linne *et al* 1986) and should find application to other gas-dynamic flows using near-IR lasers with either short-path or evanescent wave absorption.

5.2. High-speed flows

The unique requirements of high-speed flows for fast response, non-intrusive instrumentation often drive developments of new optical diagnostic techniques and diode laser absorption sensors are no exception. One of the earliest demonstrations of room-temperature diode laser sensors for gas-dynamic monitoring was based on O_2 absorption in high-speed air flows in a shock tube (Philippe and Hanson 1991, 1993). Using wavelength-modulation techniques, simultaneous measurements of temperature, density, velocity and mass flux were demonstrated over a range

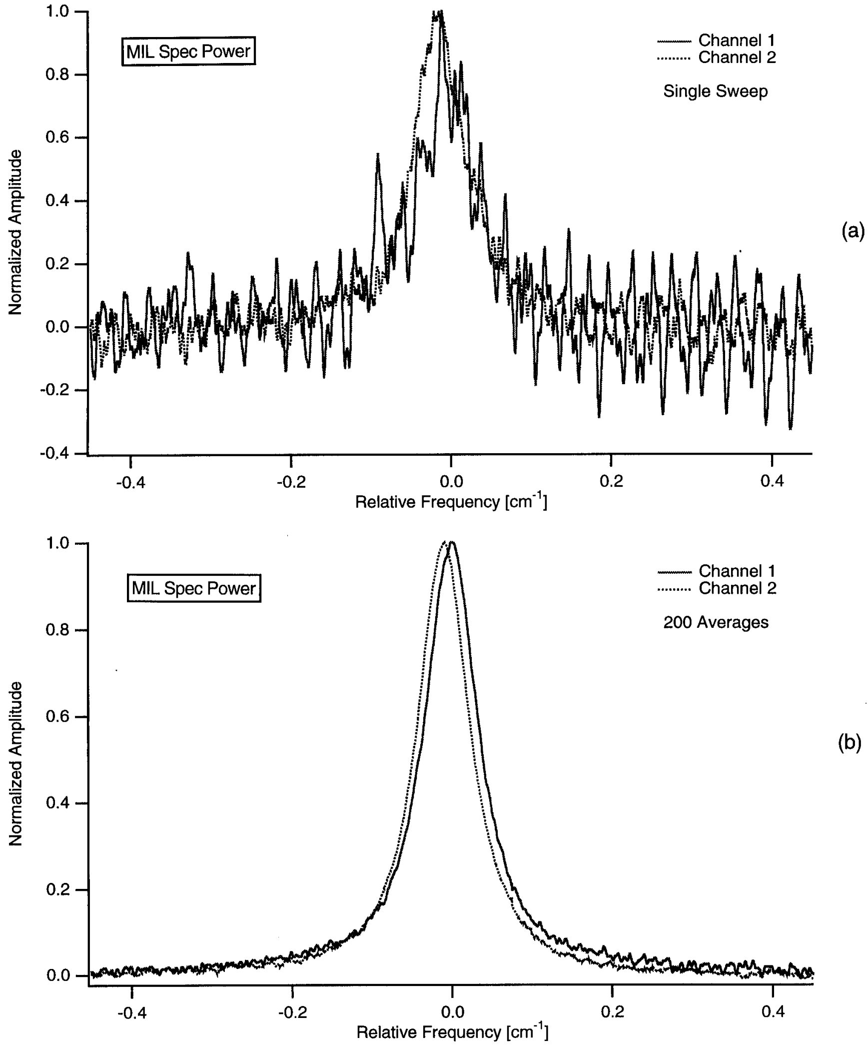

Figure 6. Example O_2 absorption lineshapes recorded in the aeroengine inlet at full (mil-spec) power conditions.

of 500–1000 m s^{-1}, 0.4–1.0 atm and 600–1100 K. The measurements demonstrated submillisecond temporal resolution. Although only a limited set of data was analysed, estimates of experimental uncertainties were 75 m s^{-1}, 0.07 atm and 75 K. This basic concept was later extended to a multiplexed system for two wavelength measurements on H_2O (thus determining the flow temperature) and O_2 for characterizing combustion in high-speed shock tunnel flows (Baer *et al* 1994, 1996, Hanson 1997).

Water vapour is a natural constituent of humid air and present in most high-enthalpy test facility flows. It is also a major product of hydrocarbon/hydrogen combustion and is thus a natural target for diode laser sensor applications. As with the O_2 measurements described above, simultaneous measurement of temperature, density, velocity and mass flux were demonstrated over comparable ranges of these parameters with comparable uncertainties (Arroyo *et al* 1994a, b). Recently, this technique has been combined with a multipass probe geometry to determine the operating characteristics of a high-enthalpy shock tunnel facility (Hanson, 1997). Figure 7 is an example from this work of a single 125 μs sweep of two lasers operating on separate transitions near 1.4 μm for a 10 MJ kg^{-1} high-enthalpy test condition. The top panel shows the Doppler-shifted absorption line recorded by a path oriented at 54° with respect to the main flow velocity and the bottom panel shows an second absorption lineshape recorded simultaneously but with a laser beam path normal to the flow velocity. The Doppler-width of each line gives the gas translational temperature, which is in good agreement with the rotational temperature inferred from the ratio of the two lines. The small peak in the top panel is from unshifted absorption due to room air and conveniently provides the frequency reference for determining the magnitude of the Doppler shift and, hence, the gas velocity of 4630 m s^{-1}. This result demonstrates the high-speed capability of laser diode frequency tuning and suggests a number of related high-enthalpy flow facility diagnostics for facility characterization and flow phenomenology.

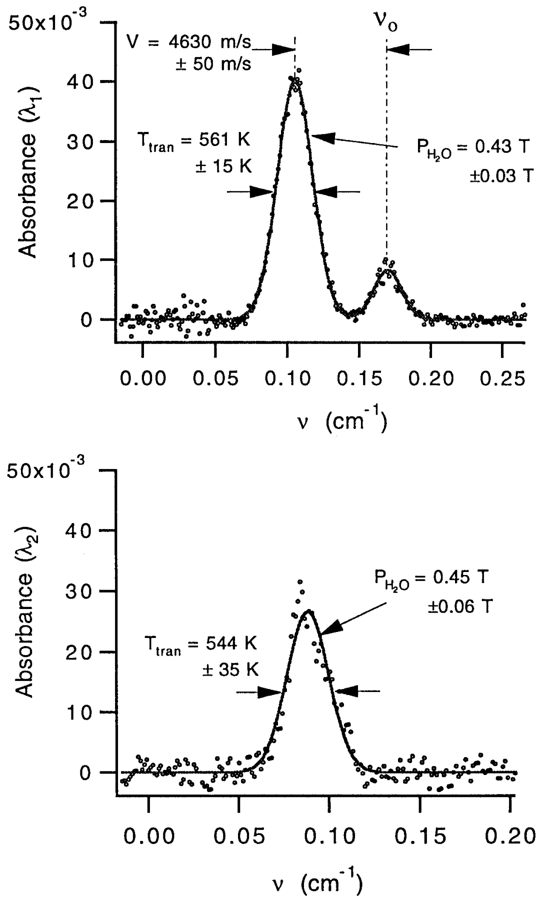

Figure 7. Example of simultaneous temperature, water vapour density, and velocity in a 10 MJ kg^{-1} shock tunnel flow field.

5.3. General flame, combustion and emissions measurements

Other than the water vapour measurements described previously, *in-situ* measurements of combustion species with room-temperature diode lasers are only beginning to appear in the literature. Initial measurements of CO, CO_2, OH, and NO have demonstrated the essential feasibility of flame monitoring, but more effort is required to understand the high-temperature spectroscopy of the targeted molecules, as well as interferences from high-temperature water vapour (Sonnenfroh and Allen 1996a, 1997a, b). Even water vapour measurements remain subject to considerable uncertainty in many bands, particularly the transitions near the technically important 1.31 and 1.55 μm bands, where the long-term availability of high-quality, inexpensive laser devices is ensured (Allen and Kessler 1996, Sonnenfroh and Allen 1996a, Upschulte and Allen 1997). Figure 8 shows a diode laser survey spectrum of several NO and water vapour transitions near 1.8 μm. The measurement was made in a cell with 74 Torr of NO in N_2 at a total pressure of 740 Torr. The neighbouring water vapour transitions arise from ambient humidity in the surrounding path and illustrate the congested nature of this spectral region for flame sensing applications. Although $\sim$30 ppm detection limits in ambient atmospheric air would appear feasible, interference from strong high-temperature water vapour lines limits the sensitivity on the transitions shown to $\sim$140 ppm (Sonnenfroh and Allen 1997b). Further work is required to identify more optimum transitions. Recent results using a cooled AlGaAsSb/InGaAsSb laser to access the stronger first overtone of NO near 2.65 μm achieved lower ambient detection limits but this wavelength region is expected to find even stronger water vapour interferences in flame environments (Oh and Stanton 1997).

Sampling the combustion gases into a low-pressure, low-temperature cell eliminates many of the interference problems associated with *in-situ* measurements. Using this approach, off-the-shelf commercial instruments have become available for combustion applications such as NH_3 monitoring in post-combustion NO_x clean-up systems for coal-fired utility boilers (Bomse 1995). Similar extraction techniques have recently been applied to sampling CO, and CO_2 in laboratory flames (Hanson 1997). Other commercially available industrial diode laser sensors include open-path monitors for HF and H_2S fence line monitoring in petroleum refineries (Bomse 1995).

An interesting example where the unique attributes of a fibre-coupled diode laser sensor enable new measurements is microgravity combustion experiments in drop towers. At a NASA Lewis facility, the entire experimental test rig falls within a 27 m shaft during the $\sim$2 s experiment. Using a multichannel fibreoptic delivery system to create multiple lines of sight, researchers have measured the water vapour distribution in diffusion flames during a drop (Silver *et al* 1995). The fibre launch optics were mounted on the drop platform while the lasers, control electronics and data acquisition system were located in a remote and fixed laboratory. Figure 9 is an example of the measured water vapour mole fraction in a microgravity propane jet diffusion flame. The two-dimensional map was constructed from six separate drops wherein the radial distribution at a fixed height was determined. The stated uncertainty in this data is between 15 and 30%, due in part to the extreme experimental difficulty of the measurement and the lack of corroborating flame-temperature measurements.

Many of the diode laser combustion sensors are being developed for eventual inclusion in aeroengine test articles where a number of photophysical and engineering challenges are being addressed. One basic issue arises from the high pressure associated with the combustor environment—typically between 10 and 50 atm. At these pressures, the collisional broadening of the absorption transition blends neighbouring transitions and complicates data interpretation since it is not possible to determine a baseline transmission by scanning the laser wavelength off the absorption line. Initial results using fixed wavelength (Nagali *et al* 1996b, Nagali and Hanson 1997) or partially resolved transitions (Allen and Kessler, 1996) at moderate pressures ($<$10 atm) and temperature ($<$500 K) are encouraging, but more work is required to extend experimental techniques to practical aeroengine combustor conditions. Work is underway in several laboratories to develop engineering solutions to optical access in combustor and engine test stands.

Successful implementation of diode laser sensors in practical aeroengine combustors may enable more sensitive

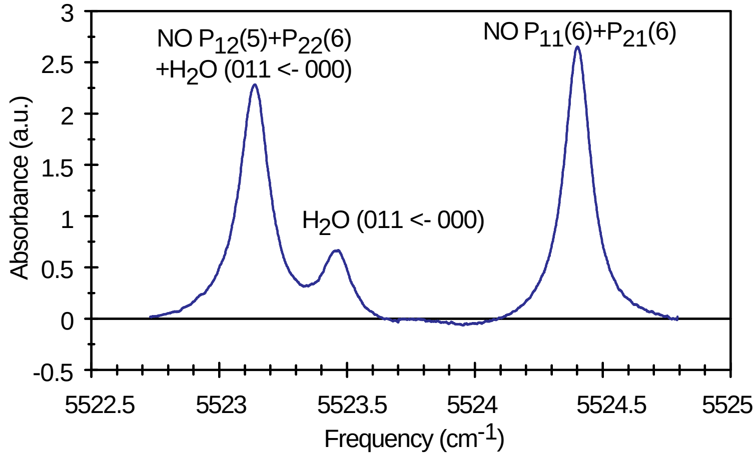

Figure 8. Example survey spectra of NO and water vapour obtained with a 1.8 μm diode laser.

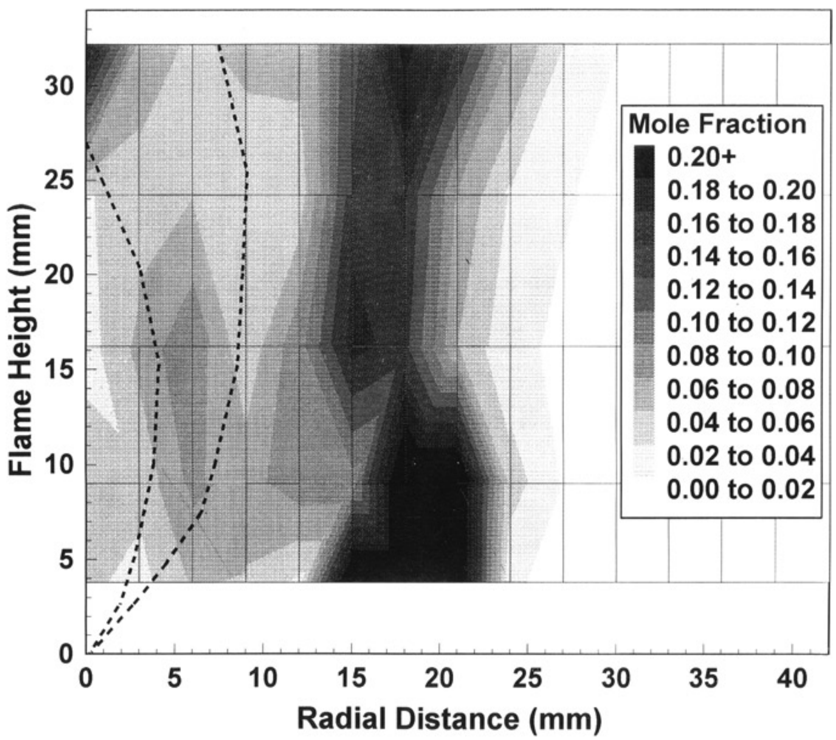

Figure 9. Example contour map of water vapour mole fraction measured in a micro gravity jet diffusion flame using tomographically inverted diode laser absorption data.

control of engine operating conditions or suppression of deleterious instabilities which limit their operational ranges. Laboratory demonstrations of prototype sensor/control systems are appearing. Using a dual wavelength multiplexed sensor for simultaneous measurements of water vapour concentration and temperature, researchers have demonstrated suppression of acoustically coupled combustion instabilities with a closed-loop bandwidth of several hundred Hz (Furlong *et al* 1997). Figure 10 is an example from this work of continuously recorded gas temperature and water-vapour partial pressure 2 cm above a flat flame burner subject to an 85 Hz instability. Since the instability is essentially one-dimensional, the path-averaged absorption data capture it well and the precision of the temperature and concentration data is on the order of a few per cent. An interesting observation made possible by this type of data is the suppression of the mean temperature and water vapour concentration, indicating that the combustion instability is reducing the overall combustor efficiency.

6. Emerging diode laser sources

Progress in the application of diode laser sensors to gas-dynamic and combustion flows continues to be driven by rapid developments in the laser-source technology and we can anticipate several areas that will have near-term impact

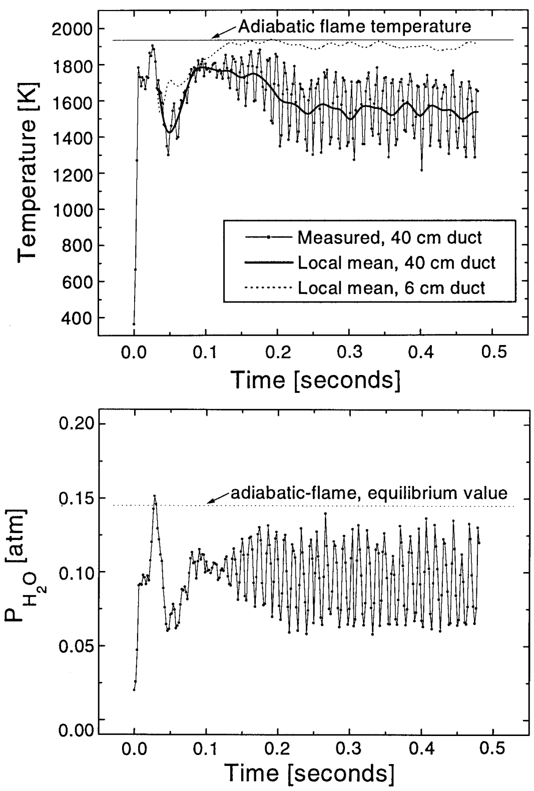

Figure 10. Measured combustion temperature (top) and water vapour concentration (bottom) in a premixed laboratory CH_4-air flat flame burner subject to an acoustic instability.

on sensor architecture and applications. Direct current injection of green and blue–green visible lasers is a major aim of the optical storage portion of the diode laser market. Progress in GaN- and ZnSe-based devices continues, but it is likely to be several years before the development of room-temperature, cw sources appropriate for sensor applications. A technology immediately available for extending the short wavelength range of diode laser sensors is nonlinear frequency conversion in KDP, BBO, or some other appropriate material. Although the conversion efficiency in conventional materials is low due to the low cw output power available from diode lasers, they are particularly interesting because they allow diode laser sources to operate in the 200–300 nm region where strong electronic transitions of species such as NO and OH are available with linestrengths 4–6 orders of magnitude larger than the near-IR transitions currently used. For example, sum frequency generation in conjunction with a fixed wavelength, high-power source has been used to detect OH at 308 nm with ppt sensitivity, despite only 1.5 μW of available laser power (Oh 1995). The recent availability of high-power, master-oscillator power-amplifier (MOPA) diode lasers near 860 nm has allowed researchers to generate deep-UV light, using mode-locking or pulsing of the near-IR source to improve the conversion efficiency for second, third, and fourth harmonic generation (Goldberg and Kliner 1995a, b). A tunable source based on fourth harmonic conversion has been used to detect NO at 215 nm with a detection limit below 1 ppm (Kliner *et al* 1997).

The efficiency of the frequency conversion may be improved using periodically poled materials in either bulk or waveguide geometries, allowing quasiphase-matched conversion over longer interaction lengths (Webjörn *et al* 1997). Frequency-doubled diode lasers in periodically poled lithium niobate waveguides have been applied to measurements of visible transitions of NO_2 (Mihalcea *et al* 1996) and, using Doppler-shift absorption techniques, as a deposition controller for Al sputtering devices (Wang *et al* 1995). A number of other materials for deep UV conversion are under development but presently suffer from gradual optical degradation at moderate laser power levels.

Alternatively, difference frequency generation in periodically poled lithium niobate or bulk $AgGaSe_2$ or $AgGaS_2$ has been demonstrated as a relatively simple approach for generating tunable radiation from room-temperature sources at wavelengths between 3.2 and 8.7 μm (Petrov *et al* 1995, Balakrishnan 1996, Eckhoff *et al* 1996, Kronfeldt *et al* 1996, Petrov *et al* 1996a, b). These lasers return room-temperature sources to the wavelength region traditionally accessed with cryogenic Pb-salt lasers. Here, strong fundamental band absorption transitions are available with linestrengths 2–3 orders of magnitude larger than the near-IR transitions described earlier. For example, ppb detection limits for CO have been demonstrated using a 4.6 μm source derived from difference-frequency mixing a diode-pumped cw Nd:YAG laser at 1.06 μm and a tunable 850 nm diode laser (Petrov *et al* 1996a). A compact, tunable spectrometer based on this technology has been demonstrated for the detection of N_2O, CO_2, SO_2, H_2CO, and CH_4 (Töpfler *et al* 1997). It seems likely that these sources may soon replace cryogenic Pb-salt lasers altogether.

These sources may in themselves soon be replaced by advances in direct-current injection devices which recover the inherent simplicity and tuning range of the near-IR and visible lasers. Progress in materials technology for InAsSb/InAlAsSb, InGaAs/InAsSb, InAsSb/InPSb, GaInAsSb/AlGaSb, and InAsSb(P)/InAsSbP devices continues to produce new mid-IR lasers in the 2–4 μm region (Wu *et al* 1997, Hasenberg *et al* 1997). Most of the devices are presently multimode and operate at near liquid nitrogen temperatures, although improvements in fabrication techniques are expected to produce cw devices operating at temperatures >250 K—accessible with thermo-electric coolers. Broad tuning capability has been demonstrated around 3.85 μm using an external cavity configuration (Le *et al* 1996). Less mature, but potentially more significant technology in quantum cascade lasers offers the possibility of broadly tunable, room-temperature sources throughout the mid-IR (Faist *et al* 1996). In these devices, quantum confinement effects are exploited to produce photon emission from sub-band transitions within the conduction band, rather than between conduction and valence bands. The past year has seen remarkable progress in these devices, including room-temperature pulsed and cw operation in the 8–10 μm region (Sirtori *et al* 1997a, b) and single-mode DFB devices near 5.4 μm (Faist *et al* 1997, Gmachl *et al* 1997).

These new mid-IR sources are likely to find their most valuable application in the area of trace-gas monitoring in ambient atmospheric environments or low-temperature flow facilities. In high-temperature combustion applications, interference from strong CO_2, H_2O, or other hydrocarbons may make sensitive detection of trace species problematic, as work with mid-IR Pb-salt lasers for *in-situ* NO detection has demonstrated (Wormhoudt *et al* 1994). Detection of CO, however, should be straightforward.

Advances in room-temperature near-IR source technology will enable more powerful sensor applications in the near future. Single-mode power, linewidth, and price/performance ratio of these devices continues to improve. Multi-electrode lasers are becoming available in fibre-pigtailed communication-style packages. These sources separate the wavelength selection, gain, and phase control regions of the laser cavity and are available in a number of different configurations (cf Ishii *et al* 1996). Injection current tuning over 100 nm around 1.5 μm has been demonstrated (Rigole *et al* 1995). Devices have been recently applied to high-sensitivity NH_3 detection (Larson *et al* 1997) and *in-situ* measurements of CO, OH, and H_2O in combustion gases (Upschulte *et al* 1998). These tuning ranges are equivalent to external cavity devices and approach that of a conventional dye laser, thereby simplifying present multilaser sensor concepts by allowing comparable wavelength coverage with a single device.

Vertical cavity surface-emitting lasers (VCSELs) are another laser structure fabricated from conventional materials that offer some promise for gas sensing application (Morgan and Hibbs-Brenner 1995, Morgan *et al* 1995b). Using AlGaAs materials, devices have been grown with laser cavities normal to the chip plane. Low and stable threshold currents operating at $>100\,^\circ$C temperature have been exploited to achieve nearly 100 nm of continuous, single-mode tuning near 850 nm. Large arrays of laser devices can also be fabricated on a single chip and application to water vapour detection has recently been demonstrated (Hovde and Parsons 1997).

7. Summary

Room-temperature diode laser absorption sensors have been developed for a variety of gas-dynamic and combustion flows. Measurements of trace and major species concentration, gas temperature, velocity, pressure and mass flow have all been demonstrated. The compact, low-cost sensors appear poised to make the transition from research instruments to standard instrumentation in a number of industrial and practical engineering test-facility applications. Mature laser devices and fibreoptic components in the near-IR are already incorporated into commercial sensors for industrial monitoring. Specialized units for gas temperature and major species monitoring, primarily H_2O and O_2, have been successfully demonstrated on full-scale aeroengines and large-scale aerospace test facilities. Additional facility integrations in the near future will further broaden the range of demonstrated applications.

As new laser devices emerge with new operating wavelength windows, additional species detection will be demonstrated, in both ambient and combustion gases. Using these sensors for closed-loop combustion control has already been demonstrated for suppressing acoustic instabilities and several efforts are underway to develop emissions sensors (CO and NO) or combustion efficiency sensors, (CO, CO_2, and O_2) to be used in combustor or engine control systems. Diode laser sensors in the mid- and near-IR have been flown on aircraft engaged in atmospheric sensing for many years, and a compact diode laser-based air mass flux sensor is under development for flight demonstration in 1998. These and related demonstrations should provide crucial data regarding the practical utility of diode laser absorption sensors for engineering research and development, as well as routine system operation.

In the next few years, integration of existing near-IR and visible sensors for O_2, H_2O, CO, CO_2, and temperature with challenging practical gas-dynamic and combustion test facilities will be an important component of progress in the field. These integrations will address problems associated with high-pressure flows, large-scale measurements, fibreoptic distribution to multiple measurement locations, high-temperature optical access, flow-quality effects, multiplexed configurations, compatibility with flight requirements and extended autonomous operation. As the sensor technology matures, such successful demonstrations will be necessary to transfer the sensors from the laser diagnostic research laboratory to the practical engineering research and development community.

The incomplete absorption database in the near-IR will continue to require substantial work in fundamental spectroscopic studies, particularly at high temperature. Broad surveys and spectral assignments are required for H_2O, CO_2, and small hydrocarbons such as CH_4 and C_2H_2 at wavelengths between 1 and 2 μm. The recent release of the Air Force HITEMP database is a major improvement for water assignments, but few validations are available and the database is only intended for use up to 1000 K. Errors in the high-temperature CO_2 database are already emerging. Extensions of this and additional databases to 2000 K are necessary for a comprehensive understanding of optimal combustor sensor strategies.

Multi-electrode near-IR devices with $\sim$100 nm tuning range are particularly intriguing for gas sensing applications. Their use in sensor configurations needs to be carefully explored in order to understand optimal methods for incorporating their broad tuning range into present sensor configurations. Because of the large market potential for dense wavelength division multiplexing in telecommunications, the availability and quality of these devices near 1.55 μm can be expected to improve considerably in the next two to three years, as with the conventional DFB devices at 1.31 and 1.55 μm. Because of this stable and growing vendor base, applications using these lasers will always have a cost and reliability advantage over custom-fabricated lasers at other wavelengths.

Frequency conversion in nonlinear materials has already demonstrated improvements in diode laser sensor detection limits by accessing stronger UV and mid-IR transitions. Commercial devices are nearing release for frequency-doubled (blue–green) and tunable difference-frequency-generated mid-IR sources. Tuning characteristics, frequency and amplitude stability and general operational parameters of these devices should receive careful attention from the diode laser sensor community. Deep UV sources (below 300 nm) continue to require improvements in the basic properties of the nonlinear materials and methods for rapid and stable frequency tuning, especially for periodically poled materials.

Despite the excitement associated with new room-temperature mid-IR sources, it is important to recognize that fibreoptic components and distribution are a major and valuable attribute of near-IR diode laser sensors and much of this advantage is lost beyond 2 μm. Fluoride, sapphire, and chalcogenide fibres are available, but not with cost and performance characteristics of telecommunication packaging. On-chip, optical waveguides carrying near-IR tunable diode laser light are expected to be incorporated directly as sensor elements for gas-dynamic and combustion applications in the near future. Long evanescent wave absorption pathlengths can be fabricated into small physical dimensions, allowing essentially point measurements from a fibre-coupled sensor (Tai *et al* 1987). Emerging technologies in micro-fabrication of electromechanical and electro-optical systems can incorporate near-IR waveguides for distributed sensing in micron-sized chemical reactors and flow devices. The transparency of the Si and SiO_2 materials used in these systems allows near-IR diode laser to penetrate structural elements for measurements in flows without special 'optical' access. This is potentially another example of a fundamental technology whose engineering development will be accelerated using sensitive, non-intrusive optical measurement techniques.

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