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Laser-Induced Fluorescence Diagnostics Using a Two-Line Excitation Method

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This paper presents recent laser-induced fluorescence measurements of oxygen temperature and density using a tunable ArF excimer laser operated in the single-pass oscillator amplification mode. The diagnostic method reported herein differs from previously reported techniques in that the temperature and density information was obtained by exciting two fluorescence transitions rather than by monitoring the Raman intensity and the fluorescence from one excitation transition. The advantages of the current method are that measurements can be obtained in flows with significant particulate loading, in flow regions near aerodynamic surfaces, and in regions of low oxygen density.

Introduction

THE Arnold Engineering Development Center (AEDC) requires nonintrusive diagnostic techniques for measurements in many of its supersonic and hypersonic ground test facilities. These measurements are needed for facility characterization and monitoring, computational fluid dynamics code validation, wake physics research, boundary- and shear-layer studies, and understanding of plume/flow interaction. The static density of these flows can range from 10^{15} to 10^{19} molecules per cm^3, and the static temperatures can range from 50 to 5000 K. For rarefied regions the electron beam fluorescence technique (EBF) is often utilized, and for the higher density regimes Raman spectroscopy is used as long as the flowfield is relatively particle free and the measurement volume is not near a surface. For almost the entire density range, laser-induced fluorescence (LIF) is a viable technique. The development of tunable excimer and Nd:YAG/dye laser systems has provided extraordinary opportunities for exciting discrete molecular transitions of many of the species in AEDC facility flows.

Because most of the flows in AEDC facilities are oxygen bearing, laser-induced fluorescence of O_2 using an ArF excimer laser, which was first proposed by Massey and Lemon,[1] appeared as a particularly worthwhile LIF technique. The O_2 Schumann-Runge band system has numerous transitions lying within the tuning curve of an ArF excimer laser, which has a nominal center wavelength of 193 nm. The O_2 fluorescence that follows excitation by the excimer laser covers the spectrum from nominally 200 to 400 nm, and the intensity of the fluorescence is a function of O_2 density and temperature. Laufer et al.[2] have shown that the LIF-O_2 technique can be used with Raman scattering to deconvolve the dependence of LIF signal on density and temperature and obtain point measurements of temperature and density. Indeed, Fletcher and McKenzie have advanced the technique to simultaneous measurement of temperature and density in a low-speed air flow[3] simulating the freestream density and temperature conditions of the Ames 3.5-ft hypersonic wind tunnel operating at Mach 5. The work in Refs. 2 and 3 has thus far been limited to temperatures ranging from 140 to 500 K and air densities from approximately 8.1×10^{18} to 5.2×10^{19} molecules/cm^3.

For three reasons, AEDC has pursued a method to supplement the Ames work. These reasons are that the temperature/density application range at AEDC is very wide, the facility flowfields sometimes have significant particulate concentrations, and many applications require measurements near solid surfaces. The AEDC method is a two-line excitation technique. It requires excitation of two different O_2 transitions, and the ratio of the subsequent fluorescence is a function of temperature and is independent of O_2 concentration. Following measurement of temperature, one of the fluorescence signals can be used to determine O_2 density using a calibration obtained at a fixed and known temperature. This method has several advantages: 1) it does not rely on Raman scattering and can therefore be applied over a wide range of conditions, in flows with particulates, and near aerodynamic surfaces; 2) transitions can be selected to optimize sensitivity to temperature; 3) molecules other than O_2 having transitions within the laser tuning range can be studied; and 4) the tuning curve can be centered at a nominal 248 nm as well as 193 nm, which gives additional flexibility. The major disadvantage to the method is that two excimer laser systems are required.

The overall objective of the AEDC work is to develop the two-line method for measurement of O_2 number density and temperature over as much of the AEDC application range as possible, and with uncertainties less than 5%. Initial demonstration and verification of the method in a laboratory quasi-static test cell is described herein.

Laser-Induced Fluorescence by the Two-Line Method

The LIF-O_2 process uses a pulsed, tunable, narrow line excimer laser to excite rovibronic transitions of the O_2 Schumann-Runge bands. The lower and upper electronic states are designated $X^3\Sigma_g$ and $B^3\Sigma_u$, respectively. As the excited molecules make downward transitions to the rovibronic levels of the ground state, the energy loss results in emitted radiation (fluorescence) in the 200- to 400-nm spectral region. Excited molecules may also lose energy through nonradiative processes. A critical feature of the Schumann-Runge system is the nonradiative process called predissociation which results from overlapping of the $B^3\Sigma_u$ energy levels with the dissociation continuum of a repulsive electronic state. The predissociation process causes most of the excited molecules to split apart rather than to fluoresce. Indeed, the ratio of the number of fluorescing molecules to the number of excited molecules is approximately 10^{-5}, and this results in a fluorescent life-

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time of a few picoseconds. This short lifetime drastically reduces the influence of collisional quenching as another nonradiative process.

The magnitude of the fluorescence signals depends on temperature and oxygen number density. To determine temperature and number density from the fluorescence signals, some method of deconvolution of the temperature and density dependence must be used. The separation technique selected is illustrated in Fig. 1. In principle, two excimer laser systems are tuned to different O_2 excitation transitions. The two laser pulses are separated in time by approximately 100 ns, and this serves to provide practically instantaneous fluorescence signals S_F relative to the time scale of molecular processes in the flow. As shown in Refs. 2 and 3, the fluorescence signals will depend linearly on O_2 number density as a result of the small influence of collisional quenching. Therefore, a ratio of fluorescence signals from these two laser pulses will be independent of O_2 number density and will depend only on temperature. The temperature sensitivity of the ratio can be optimized with selection of various excitation transitions, and calibration of the ratio variation with temperature yields a means of determining temperature in a flowfield. A calibration of the density dependence of the fluorescence signal from one of the excitation transitions at a fixed, known temperature coupled with calibration of the temperature dependence of the signal from the same excitation transition at a fixed, known O_2 number density provides a means for determination of density in a flowfield.

A computational model (Code LIFOXY) of the LIF-O_2 process was developed to aid in the selection of optimum experimental conditions, to compare to experimental results, to extrapolate beyond the bounds of calibration, and to analyze all of the results. The mathematical description of the excitation/emission process for LIF-O_2 was taken from Ref. 3. Absorption transitions from the first six vibrational levels (each with 40 rotational levels) of the ground electronic state can be calculated. Additional processes such as Raman, Rayleigh, and Mie scattering; laser-induced incandescence of particulates; and temperature perturbation were included in the code. The laser pulse was assumed uniform in space and time, and the laser line spectral profile was modeled as a Voigt function. The O_2 excitation transition profiles were also modeled as a Voigt with Lorentzian contributions from predissociation broadening and pressure broadening and the Gaussian contribution from Doppler broadening. Although the predissociation phenomenon minimizes the influence of collisional quenching, it does not make it negligible at near atmospheric densities. Therefore, a quenching rate taken from Miles et al.[4] was included in the code. An option was included to model the broad background from the laser if it is not operating in a 100% locking efficiency mode. A rectangular, Gaussian, or trapezoidal spectral profile can be selected. Spectral constants and parameters were taken from current literature. Output from the code can be in terms of excitation spectra, emission spectra, and optical filter throughput with selection of either photomultiplier or intensified array detection. Spectral sensitivity curves for a variety of spectrometers and gratings are included for optional use.

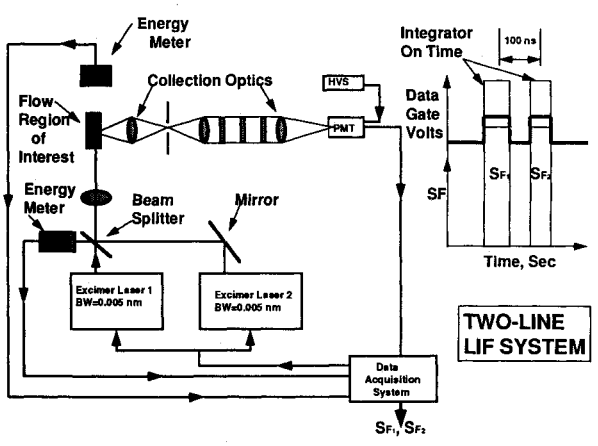

Fig. 1 Two-line LIF-O_2 method.

Code LIFOXY was first used to select two transitions that could provide a fluorescence intensity ratio with good sensitivity in the 300- to 500-K temperature range of an existing laboratory quasistatic cell. The two absorption transitions selected were the P15 ($v'' = 0$) and the P23 ($v'' = 1$).

Experiment

The excitation source was a Lambda Physik EMG-150 TMSC ArF excimer laser. The amplifier optics were removed and the laser was operated in the single-pass oscillator amplification mode. This ensured that only narrow bandwidth (0.005 nm) radiation would be emitted, permitting energy normalization of the resulting fluorescence signal. In the standard amplifier amplification mode, 10% or more of the laser energy is present in the form of broadband output, resulting in the excitation of additional transitions. The only disadvantage of operating in the oscillator amplification mode is the low output energy, normally only 1–2 mJ, compared with approximately 100-mJ output in the amplifier amplification mode. The energy output was increased approximately fivefold (8–9 mJ) by increasing the amount of energy extracted from the oscillator as described by Wodtke et al.[5] This was achieved by replacing the standard 1-mm-diam apertures at the front and rear of the oscillator with 1×5-mm slits. The beam was then compressed to a 1×1-mm square with a 5X reducing cylindrical telescope consisting of two 0.5-in.-diam cylindrical lenses, of focal lengths 20 and 100 mm, separated 120 mm. The cylindrical lenses were antireflective coated on both sides to reduce insertion losses. The telescope ensured that the beam would be injected along the amplifier axis and also allowed easy return to the normal mode of operation with little more than a replacement of the amplifier optics. To facilitate tuning from one transition to the other, the manual grating drive control was replaced with an electronic servocontroller. To exclude the strongly absorbing O_2 from the beam propagation path, it was necessary to purge the entire path with nitrogen gas. This was accomplished by replacing the original optics table covers with a system of new covers and window seals. The beam stability and charge lifetime of the laser were greatly enhanced by constructing a cryogenic gas purification system consisting of a gas circulation pump, particle filters, and a LN_2 cryotrap. This improvement also allowed operation for as long as 4 days with only daily injections of small amounts of He/F_2 gas mixture.

The test cell consisted of a small chamber with two viewing windows and one laser entrance window made of thin MgF_2. The laser energy density was approximately 0.8 J/cm^2. The buildup of ozone and other laser generated byproducts was prevented by slowly flowing the test gas (bottled breathing air with 0.01-μm particle filtration) across the focal volume. A thermocouple located approximately 0.25 in. above the focal volume indicated the test gas temperature. A Baratron transducer was used for pressure measurements. It was verified that the temperature and pressure 0.25 in. above the focal volume were the same as at the focal volume. The fluorescence signal was coupled to a solar-blind photomultiplier detector system (RCA C31034A with Corion SB-300 filter) via a single fused silica lens viewing a 1-mm^3 focal volume. Emission spectra were dispersed with a 0.275-m monochromator coupled to an RCA C31034A photomultiplier tube (PMT). The data were acquired with an LSI-11 based gated integrator system. Fluorescence signals were energy normalized to eliminate pulse-to-pulse fluctuations.

Since only one laser was available, it was necessary to record the fluorescence information from the P15 ($v'' = 0$) transition and then tune to the P23 ($v'' = 1$) transition. This was accomplished by connecting the PMT system output to a boxcar averager and tuning the laser to the desired transition by ob-

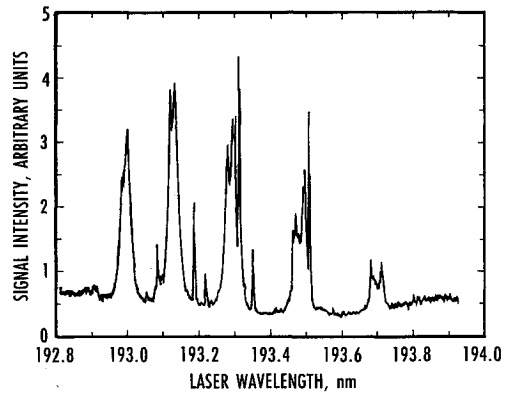

Fig. 2 Typical O_2 excitation spectrum prior to improvements.

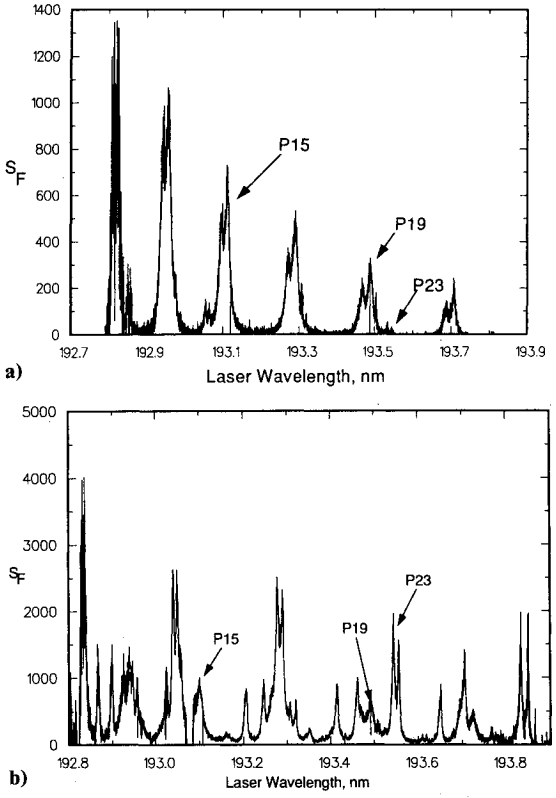

Fig. 3 Excitation spectra after improvements: a) $T = 297$ K, density $= 6.53 \times 10^{18}$ molecules/cm^3; b) $T = 500$ K, density $= 6.53 \times 10^{18}$ molecules/cm^3.

serving the spectra on a digital oscilloscope. The test cell conditions were held constant throughout the tuning process.

Experimental Data

The performance of the laser prior to the incorporation of the improvements just described is illustrated in Fig. 2, which is a typical O_2 excitation spectrum at a temperature of 300 K and a pressure of 100 Torr. Note the high baseline on both the lower and upper edge of the tuning range, indicating poor locking efficiency. The narrow multiphoton emission lines are from O_2+ generated because the fluence level was too high (>1.5 J/cm^2) (Refs. 2 and 3). The performance of the improved laser is evident upon examination of Figs. 3a and 3b, which illustrate energy normalized constant density (6.53×10^{18} molecules/cm^3) excitation spectra at 297 K and 500 K, respectively. The drastic increase in the P23 ($v'' = 1$) line signal strength in comparison to the P15 ($v'' = 0$) and P19 line signal strengths clearly shows the reason for the choice of these two transitions for this temperature range. Note also the flat baseline over the extent of the tuning curve, and the highly resolved peaks.

Figures 4a and 4b show the excellent agreement with theory, as predicted by the LIFOXY code. The coincidence of the two spectra over virtually the entire tuning curve, and the flat baseline are evidence of 100% locking efficiency. The slight wavelength shift of the experimental data in Fig. 4b was due to a nonlinearity in the laser scan control drive mechanism. (Note

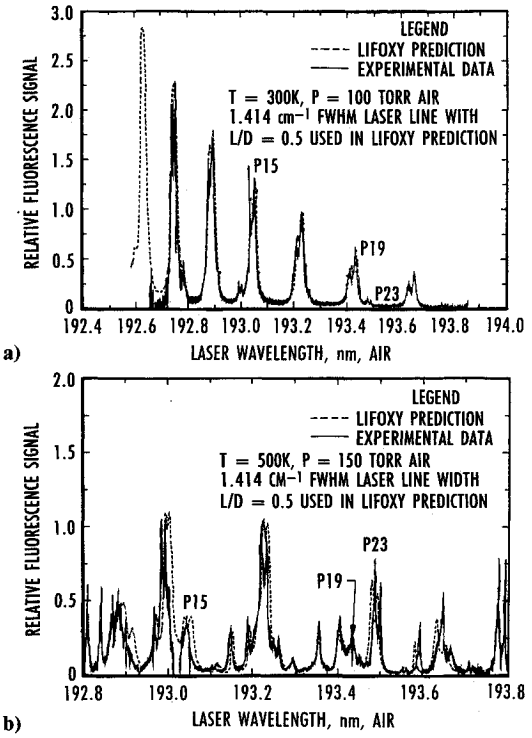

Fig. 4 Comparison with theory: a) $T = 300$ K and b) $T = 500$ K.

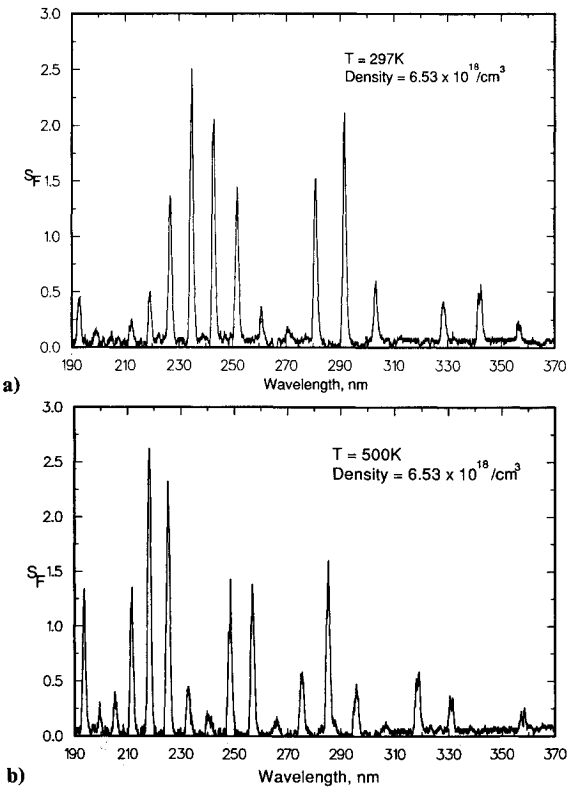

Fig. 5 O_2 emission spectra: a) P15 excitation, $T = 297$ K; b) P23 excitation, $T = 500$ K.

that L/D is the ratio of the Lorentzian component width to Gaussian component width, used in generating the LIFOXY spectra.)

Figures 5a and 5b show the emission spectra resulting from the excitation of the P15 ($v''=0$) transition at 297 K and the P23 ($v''=1$) transition at 500 K, respectively. The intensities of the two transitions, P23 ($v''=1$) and P15 ($v''=0$), were recorded over a range of temperatures (300–500 K) at constant density for three densities: 1.76, 2.69, and 4.04×10^{18} mole/cm^3. The results of these measurements are shown as constant density plots of the ratios of the P23 ($v''=1$) and P15 ($v''=0$) signals in Fig. 6. Each symbol on the curve represents the average of 500 energy normalized pulses. As illustrated, the resultant curves were found to be virtually density independent. A temperature calibration curve was developed by averaging the three constant density curves.

The P15 ($v''=0$) line was used to determine density. A density calibration factor was determined from measurements of signal strength at 300 K and a known low density. As previously shown, the P15 ($v''=0$) line is much less temperature sensitive than the P23 ($v''=1$) line, but the slight temperature sensitivity must be taken into account to accurately determine the density. Once the temperature is determined from the P23 ($v''=1$) and P15 ($v''=0$) signal ratio, the corresponding density is determined by applying the density calibration and temperature correction factors. A collisional quenching correction factor was then applied using LIFOXY.

Data were acquired at densities ranging from $1.50\text{–}8.75\times10^{18}$ molecules/cm^3, and temperatures of 300–550 K. The results of the measurements are shown as a temperature-density plot in Fig. 7. Note that in most cases the measurement errors are within ±5% of the thermocouple and pressure transducer measurements.

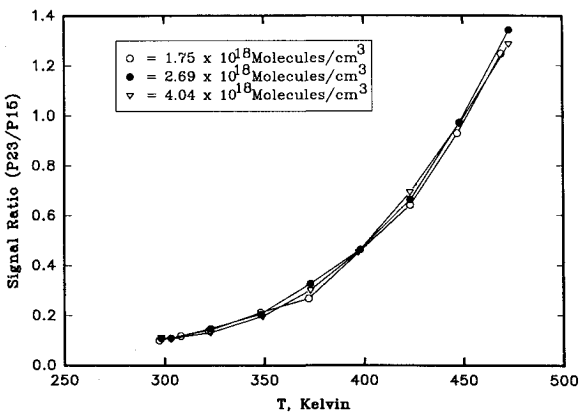

Fig. 6 Ratio of P23 and P15 signals at three densities.

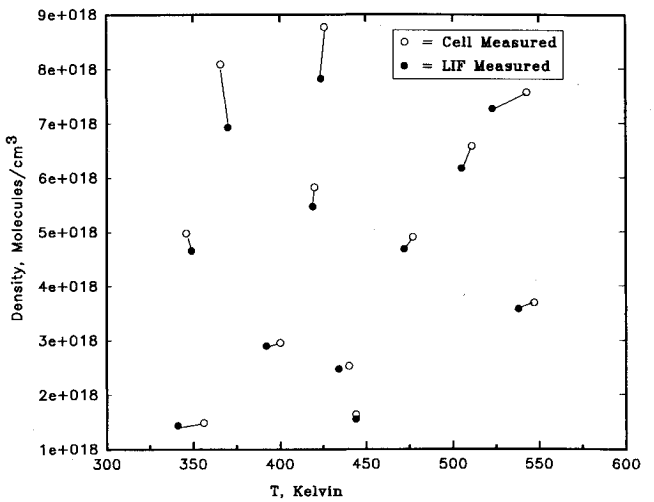

Fig. 7 Comparison of LIF and thermocouple/pressure gauge measured temperatures and densities.

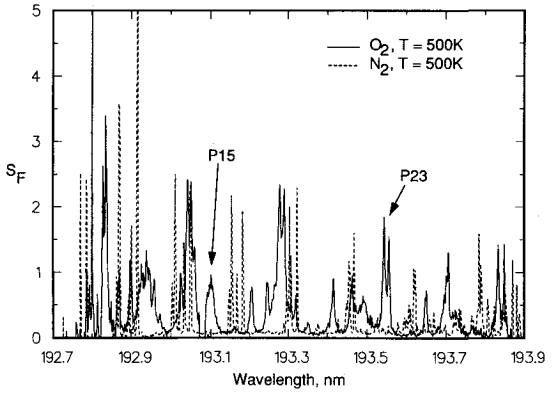

Fig. 8 Overlay of O_2 and NO excitation spectra.

Certain test conditions are such that nitric oxide (NO) can be present in significant quantities. For this reason, experiments were performed to determine if the use of the P15 ($v''=0$) and P23 ($v''=1$) absorption transitions for O_2 could produce excitations and interfering fluorescence from the NO beta or epsilon bands. Figure 8 shows an overlay of the excitation spectra of the O_2 Schumann-Runge band and the NO beta and epsilon bands at a temperature of 500 K. Note that the P15 ($v''=0$) and P23 ($v''=1$) transitions would produce fluorescence relatively free from NO interference. This would not be possible if the laser were operating in the standard amplifier amplification mode because the broadband output would excite nearby transitions.

Summary

The objective of the present AEDC work has been to develop the two-line LIF method for measurement of O_2 density and temperature over as much of the AEDC application range as possible. The initial demonstration and verification of the method in a laboratory quasistatic test cell using a tunable ArF excimer laser has been completed. The experiments were successfully guided by a computational model (Code LIFOXY) developed for the LIF-O_2 process. Comparison of code predictions with experimental results indicates that the code will be an accurate tool for future work. The experimental results also indicate that measurement errors less than ±5% are achievable. A complete uncertainty analysis is currently in progress.

In keeping with the overall objective, additional laboratory measurements are underway using the same excimer laser system set up for KrF operation at a nominal 248 nm. Recent work by Laufer[6] has indicated that for the temperature range from approximately 500 to 1800 K, the KrF laser is the best choice. Furthermore, based on AEDC work with NO and the recent work of Lempert et al.[7] with H_2 using the ArF system, the scope of the AEDC work is being expanded to include measurements of NO and H2 density. Validation experiments will be performed in both a resistance heated supersonic flow and a combustion driven high-enthalpy test bed.

Acknowledgments

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