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ASYNCHRONOUS OPTICAL SAMPLING FOR LASER-BASED COMBUSTION DIAGNOSTICS IN HIGH PRESSURE FLAMES

Final Report Air Force Office of Scientific Research Grant No. AFOSR-84-0323 December 15, 1987 - January 31, 1989

Galen B. King^{*}, Normand M. Laurendeau^{*}, and Fred E. Lytle[†] *School of Mechanical Engineering [†]Department of Chemistry Purdue University West Lafayette, IN 47907

Abstract

This report describes progress on the development of a new laser-based combustion diagnostic for the quantitative measurement of both major and minor species in high pressure flames. The technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond pump/probe spectroscopy. The timing parameters for the current ASOPS instrument are described and consideration is given to the optimization of these parameters. Results from the study of the sodium atom in an atmospheric flame are presented. Instrumental changes are shown that considerably improve the ASOPS signal-to-noise ratio. A technique for optimization of ultra-violet generation is demonstrated and initial hydroxyl fluorescence experiments are described.

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1. RESEARCH OBJECTIVES

The overall goal of this research is to develop and test a new combustion diagnostic for the quantitative measurement of both major and minor species in high pressure flames. The proposed technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond laser spectroscopy which should yield a better signal-to-noise ratio than laser fluorescence measurements in rapidly quenched combustion environments. Furthermore, ASOPS will allow determination of both quenching rates and state-to-state relaxation rates which are necessary for quantitative applications of both laser-induced and laser-saturated fluorescence at high pressure. The ASOPS technique produces a coherent signal-carrying beam and thus requires no more optical access to practical combustion devices than LDV measurements. By applying the proposed method, "real-time" concentrations of important species such as OH, NO and CO can be measured in flames at 1-20 atm.

2. RESEARCH STATUS

2.1 ASOPS Methodology

Absolute number density measurements in flames are difficult to obtain from laserinduced fluorescence (LIF) signals because the rate of fluorescence quenching due to collisional deexcitation is not well known.¹ Laser-saturated fluorescence (LSF) can be used to avoid the quenching dependence² but this technique is limited by the number of mc is ules that can be effectively saturated. Thus, in flames above 1 atm, measurement of number density requires that picosecond lasers be used to directly determine the rapid quenching rates. Previous investigators have used picosecond lasers to do time-resolved LIF experiments on OH at atmospheric pressure using either a streak camera^{3,4} or a time-correlated single-photon counting scheme.⁵ Although these experiments have the temporal resolution necessary to obtain the needed quenching rates, the amount of time necessary to obtain the data is much greater than the time scale of turbulence.

Because of these limitations, we have sought and developed a new laser-based combustion diagnostic for turbulent, high-pressure flames. The technique, Asynchronous Optical Sampling (ASOPS), is a pump/probe method; however, it overcomes many of the temporal problems inherent to that method ^{6,7} Furthermore, corrections for the effects of quenching can be obtained on a time-scale necessary for practical combustion measurements.^{8,9} In the ASOPS method, rather than using a single Nd:YAG laser to construct both the pump and probe beams with an optical delay line to control the relative timing between the two pulses, two Nd:YAG lasers are used to separately generate the pump and probe beams. Central to the method is the fact that the two Nd:YAG lasers are mode-locked at slightly different frequencies. The mode-locking frequencies of these two lasers are carefully controlled to maintain a constant beat frequency, creating a periodic relative phase walk-out between the pump and the probe lasers. This has the same effect as varying the optical delay line in the conventional pump/probe method; however, the period of time needed to observe the population decay of an excited energy level can be reduced from several minutes to less than a msec.

The process is illustrated in Fig. 1a, which shows the excited state population produced by several pump pulses and the temporal position of several probe pulses. Each successive probe pulse is delayed in time (relative to the pump pulse train) by a constantly increasing duration which is determined by the beat frequency of the system. Thus, each probe pulse samples the excited state population at a slightly later time than the preceding probe pulse. This is equivalent to varying the optical delay in a conventional instrument. The sampling process repeats itself when the cumulative delay equals the period of the pump laser. Hence, any modulation of the probe beam, resulting from the creation and subsequent decay of the excited state, repeats at the beat frequency of the system. Therefore, in contrast to a conventional pump/probe instrument, there is no need to modulate the amplitude of either beam to employ synchronous detection.



Fig. 1b illustrates the change in probe intensity which occurs upon stimulated emission from the excited state population shown in Fig. 1a. The net effect of the ASOPS technique is that a small amplitude waveform, which is directly related to the fluorescence decay of the species under study, is impressed onto the probe laser intensity. In essence, a temporal transformation of the excited state decay is performed with the time scaled by the factor ($f_{pump}/(f_{pump} - f_{probe})$) where *f* is the repetition rate of the two lasers. The ASOPS technique is thus an optical analogue of the sampling oscilloscope.

Since the wavelengths of both the pump and the probe lasers can be independently controlled, specific electronic quenching rates between the excited and the ground states can also be determined. This assertion is valid because the stimulated emission (absorption) appears as a gain (loss) in the probe laser intensity; therefore, only those states which are directly connected by the probe laser wavelength are sampled. The loss/gain in the probe laser beam also provides information on the rate at which the depopulated lower level gains population from neighboring levels within the ground electronic state, or the rate at which the populated upper level transfers population to its neighboring levels within the excited electronic state. In other words, for molecular species the ASOPS technique can yield information about state-to-state relaxation rates within the upper and the lower rovibronic manifolds.

The beat frequency of the ASOPS instrument is determined by the difference in laser repetition rates or $f_{\text{beat}} = f_{\text{pump}} - f_{\text{probe}}$. The period of the pump laser, $1/f_{\text{pump}}$, determines the free temporal range, which is the maximum time available for a single decay of the excited state population. The inverse of the beat frequency, $1/f_{\text{beat}}$, represents the collection time for a single decay. The total number of points sampled during the decay is determined by $f_{\text{probe}}/f_{\text{beat}}$. The temporal difference between each sampled point within the decay profile is the sampling interval. The sampling interval and thus the temporal resolution is determined by the difference in laser periods, i.e., $f_{\text{beat}}/(f_{\text{probe}} f_{\text{pump}})$. Since the ASOPS signal follows a simple exponential, the

duty cycle of the measurement is determined by the excited state lifetime of the species being monitored and the free temporal range. If we assume that the signal is zero after 3τ , where τ is the excited state lifetime, then the percent duty cycle is given by $100(3\tau f_{pump})$.

Table 1 gives the above operating parameters for various beat frequencies and laser repetition rates. Several factors must be taken into account to determine the optimum ASOPS parameters. First, to make measurements on the time-scale of turbulence, it is necessary to collect enough data for adequate signal averaging in \sim 1 msec. This criterion indicates that we should use the largest possible beat frequency. However, the beat frequency is limited by the physical cavity length of the lasers and the natural acoustic resonance frequencies of the mode-locking prisms. In addition, since the ASOPS signal is carried on the probe beam, the beat frequency must be chosen so as not to coincide with any noise in the frequency spectrum of the laser.

A second factor important to system optimization concerns the sampling interval, which must be small enough so that the ASOPS measurement has sufficient temporal resolution to reconstruct the excited state decay. This criterion sets a natural upper limit to the beat frequency because the sampling interval is proportional to f_{beat} . Finally, the duty cycle should be as close to 100% as possible. As previously shown, the duty cycle increases with the pump laser repetition rate and is independent of the beat frequency. The pump laser repetition rate can be increased by a technique called third-harmonic mode-locking. As can be seen from Table 1, this approach would provide the ideal case for ASOPS. The increased repetition rate not only improves the duty cycle, but also allows for higher beat frequencies and thus SNR because of the complementary effect of f_{pump} on the sampling interval.

Table 1. OPERATING PARAMETERS FOR VARIOUS BEAT FREQUENCIES								
Repetition Rate (fpump)	~82 MHz	~82 MHz	~246 MHz					
Beat Frequency (f _{beat})	10 kHz	100 kHz	1 MHz					
Free Temporal Range (1/f _{pump})	12.2 nsec	12.2 nsec	4.1 nsec					
Collection Time (1/f _{beat})	100 μsec	10 μsec	1 μsec					
Samples per Decay (f _{probe} /f _{beat})	8,200	820	246					
Sampling Interval (f _{beat} /f _{probe} f _{pump})	1.5 psec	15 psec	17 psec					
Percent Duty Cycle (for $r = 1$ nsec)	25%	25%	74%					

2.2 Progress to Date

In the past year, we have made considerable progress with respect to both the development of the ASOPS instrument and application of the technique to a flame environment. We first describe the components and configuration for ASOPS, as employed for our measurements of atomic sodium in an atmospheric premixed flame. Improvements in the instrument have resulted in a vastly enhanced signal-to-noise ratio (SNR). Work has also been done to maximize the UV power for subsequent studies of the hydroxyl radical. The bandwidth of the laser has been reduced and initial fluorescence studies of the hydroxyl radical in an atmospheric flame have been performed.

2.2.1 Instrumental Developments

Since the ASOPS technique requires that the pump and probe lasers operate at slightly different repetition rates, the instrument must be constructed from two independent mode-locked laser systems. Both the pump and probe beams are currently derived from Spectra-Physics model 375B dye lasers, which are synchronously-pumped by frequency-doubled, mode-locked Spectra-Physics series 3000 Nd:YAG lasers. The mode-locking frequencies are

generated by two Programmed Test Sources (PTS) model 160 frequency synthesizers (accurate to 0.1 Hz) operated in a master-slave (i.e., phase-locked) configuration to minimize drift in the beat frequency of the system. Both the pump and probe beams consist of an ~82 MHz train of pulses, tunable (using Rhodamine 6G) from 56C to 640 nm. Three plate Lyot (birefringent) filters and ultrafine etalons placed in the dye laser cavities result in ~20 psec pulses with nearly transform limited bandwidths of 30 GHz¹⁰ ($\Delta\lambda = 0.04$ nm) for both the pump and probe lasers. Throughout the center of the tuning range (560 - 640 nm), average powers in excess of 120 mW are obtained using 800-900 mW of pumping power.

A block diagram of the basic ASOPS instrument is shown in Fig. 2. To obtain the trigger signal, a synchronous voltage output from each synthesizer is first amplified with an RF power amplifier (Electronic Navigation Industries 503L), frequency doubled by an electronic frequency-doubler (Anzac DI-4), and then electronically mixed by a double-balanced mixer (Anzac MDC-161). The mixer output $_{+}$...sses through a 4LM5-3-CD Texscan 5-MHz, low-pass filter to remove any high frequencies leaking through the mixer. The output of the filter then enters a digital delay generator (Berkeley Nucleonics Corporation 7095) which produces a very stable 0 to +5 V trigger pulse of variable width. Unlike the optical triggering scheme previously employed,7.11 this new electronic scheme offers much less temporal jitter in the trigger pulse and thus an improved SNR.

The pump and the probe beams are first passed through calibrated neutral density filters which provide a convenient means of varying the power in either beam. The two beams then pass through a single focusing lens (f = 100 mm) and cross at an included angle of $\sim 5^{\circ}$ in the flame. The probe beam is detected using the circuit of Fig. 3. Here, approximately half of the probe is split off before the flame and reaches one of the photodiodes (EG&G FND-100Q). After the flame, the probe is recollimated and monitored by the second photodiode. Reduction of probe-beam noise of up to 20 dB results from the high common-mode rejection ratio of the



Fig. 2. Block diagram of the ASOPS instrument.



AD 521 instrumentation amplifier. The output of the detection circuit is filtered with a 4LM5-3-CD Texscan 5-MHz low-pass filter to remove the high frequency noise. The output from the filter is amplified by a C-COR 4375-A wideband AC amplifier by a factor of 100. The output from the amplifier is then directed to a digitizing oscilloscope (Hewlett-Packard 54100A), triggered at the beat frequency of the system. Currently, the ASOPS repetition rates are $f_{pump} =$ 81.59358620 MHz and $f_{probe} = 81.58362200$ MHz, giving a beat frequency $f_{beat} = 9.964$ kHz. The interaction volume of the pump and the probe beams is ~90 pL in the flame.

2.2.2 Experimental Results

As the first substantive test of ASOPS in a combustion environment, we have chosen to detect atomic sodium that has been atomized into a premixed $C_2H_4/O_2/N_2$ flame. The flame is supported by a water-cooled, 31-mm diameter Hastelloy X flat flame burner. A NaCl solution is directed with a multistatic pump to a nebulization chamber, after which mist droplets are carried to the burner via the combined O_2/N_2 flow. Fig. 4 shows the ASOPS signal for sodium in an atmospheric flame. The pump beam is set to the sodium $D_2 (3S_{1/2} \rightarrow 3P_{3/2})$ transition (589.0 nm) while the probe beam is set to the $3P_{3/2} \rightarrow 5S_{1/2}$ transition (616.1 nm). Thus, the ASOPS signal monitors the population of the $3P_{3/2}$ state, which modulates the probe beam intensity through excited-state absorption.

The decay curve in Fig. 4 was obtained by averaging over 256 separate single-decay curves. For alkali atoms such as sodium, the decay of the $3P_{3/2}$ state is not a true exponential, but is governed by a second-order decay incorporating the quenching rate ($3P_{3/2} \rightarrow 3S_{1/2}$) and the doublet mixing rates ($3P_{3/2} = 3P_{1/2}$).^{12,13} A curve fit to the data with a .05% standard error is shown in Fig. 5, resulting in a $3P_{3/2} \rightarrow 3S_{1/2}$ quenching time of 830 psec and a $3P_{3/2} \rightarrow 3P_{1/2}$ mixing time of 500 psec. This compares to lifetimes of 710 psec and 330 psec reported by Takubo *et al.*¹³ for sodium in a propane-air flame.



Fig. 4. Temporal decay for atomic sodium obtained with a 589.6-nm pump beam (120 mW) and a 616.1-nm probe beam (3 mW). The lower scale gives the equivalent time displayed on the oscilloscope, while the upper scale gives the actual time.



Fig. 5. Second order curve fit to the decay of Fig. 4. A $3P_{3/2} \rightarrow 3S_{-2}$ quenching rate of 1.2 x 10⁹ sec⁻¹ and a $3P_{3/2} \rightarrow 3P_{1/2}$ doublet mixing rate of 2.0 x 10⁹ sec⁻¹ result in a fit (indicated by the solid curve) with .05% relative error from the experimental results (data points indicated by circles). To distinguish the curve fit from the experimental data, only every fifth data point is plotted.

The results of Fig. 4 represent a factor of 2.5 improvement in peak signal-to-noise ratio over initial measurements using the ASOPS technique. This takes place despite the 2056 samples required to obtain the initial results, in comparison to the 256 samples of the present data. Furthermore, the pump and probe beams of the initial results were tuned to the D₂ transition, which has an Einstein coefficient for absorption that is approximately 12 times larger than that of the $3P_{3/2} \rightarrow 5S_{1/2}$ transition in resonance with the probe beam in the present experiments. The resulting increase in SNR by a factor of ~100 is attributed to the new trigger arrangement, the detector of Fig. 3, and improved dye laser operation.

2.3 Current Research

Since transitions for the hydroxyl radical are in the ultra-violet region of the spectrum, much work has been done in the area of second harmonic generation. Generation of highpower, wavelength-tunable UV picosecond laser pulses is commonly believed to require the use of pulse compression and cavity dumping¹⁴, or second harmonic generation in either an intracavity geometry^{15,16} or an external-ring resonator¹⁷. These approaches use either high peak power or high circulating power to increase the generated UV. However, the addition of a pulse compressor increases the laser linewidth, while the use of a cavity dumper reduces the repetition rate of the laser. Both increase the cost and complexity of the laser system. Intracavity frequency doubling requires modification of the dye laser optics, which slaves the dye laser to UV operation and makes scanning the wavelength difficult. Moreover, the majority of such intracavity designs utilize ring dye lasers. External ring resonators require additional optical components, and critical matching of the resonator length to the dye laser cavity length must be maintained.

In our experimental arrangement, the output of the dye laser passes through a focusing lens, a LilO₃ crystal, a collimating lens and a Pellin-Broca prism. The UV power is monitored using a pyroelectric radiometer (Laser Precision Corp. Model RkP 545). The 10x10x2-mm LilO₃

crystal, cut at an angle of 60°, is encased in a cell with AR coated windows. The crystal and cell, which is filled with index-matching fluid, are obtained from Quantum Technology Inc., Sanford, Florida. A combination of translation and rotation stages allows positioning of the crystal to maximize the UV output.

Using 3.0 psec pulses at 600 nm and an average fundamental power of 390 mW passing through a 160 mm focal-length lens, 12 mW of average UV power was produced at 300 nm. The power could be increased to 28 mW with the use of a 120 mm focal length lens but the UV power dropped and then stabilized at 19 mW over a ten-minute period. This results in a conversion efficiency of between 3% and 7%, which is a substantial improvement over a reported conversion efficiency of 9x10⁻³ %¹⁶. At 635 nm the average fundamental power dropped to 185 mW and this produced 2.1 mW of average UV power at 317.5 nm. Therefore at the edges of the dye tuning curve, the conversion efficiency remained higher than 1%.

Our work in pump/probe spectroscopy^{7.18} has led us to the realization that the cavity length of the dye laser is a critical parameter in determining signal quality, and that an autocorrelation trace lacks the resolution required to determine the optimum cavity length. We have also found that the cavity length is critical in achieving the maximum conversion efficiency to the second harmonic. Cavity length changes have a pronounced effect on the UV power produced for a given fundamental power. For example, a 2 μ m change from the optimum cavity length yields a 27% drop in UV power. Hence, while an autocorrelator may be used as a diagnostic for coarse cavity length adjustment, the optimum cavity length in this case is best determined by monitoring and maximizing the UV power. Our observation is not an isolated event; comparable results were independently reproduced by other research groups under our direction using identical LilO₃ crystals on a similar laser system.

Much concern has been expressed by the flame diagnostics community that the bandwidth of our picosecond lasers would be too large to resolve the hydroxyl transitions. To investigate this presumption, fluorescence scans have been performed. As noted above, the

placement of etalons in each dye laser has reduced the laser bandwidth to ~30 GHz ($\Delta\lambda = 0.04 \text{ nm}$). This has allowed the Q₁(4), Q₁(5), P₁(2), Q₁(6), and the Q₁(7)/Q₂(3)/Q₂(2) combination to be resolved with our lasers. An absorption measurement of the Q₁(5) transition resulted in a number density of 1 X 10¹⁶ cm³, in good agreement with past experiments with similar burners. A vertical fluorescence profile is shown in Fig. 6. The exciting transition is the Q₁(7)/Q₂(3)/Q₂(2) combination.

2.4 Future Work

Future work with the ASOPS instrument will involve obtaining an ASOPS signal of the hydroxyl radical. A liquid solution and atomic copper in a flame environment may be employed to find and optimize the ASOPS signal. Atmospheric premixed flames will be used to first determine absolute number densities and quenching rates for the hydroxyl radical. A transient digitizer will be employed to upgrade the data acquisition system, thus taking full advantage of the inherent speed of the ASOPS measurement.

3. PUBLICATIONS AND PRESENTATIONS

The following ASOPS papers have been published, accepted, or submitted for publication.

- P. A. Elzinga, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, "Pump/Probe Spectroscopy by Asynchronous Optical Sampling", Appl. Spectrosc. 41, 2 (1987).
- P. A. Elzinga, R. J. Kneisler, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, "Pump/Probe Method for Fast Analysis of Visible Spectral Signatures Utilizing Asynchronous Optical Sampling", Appl. Opt. 26, 4303 (1987).
- 3. G. J. Fiechtner, G. B. King, N. M. Laurendeau and F. E. Lytle, "Determination of Relative Number Densities and Quenching Rates of Sodium in an Atmospheric





Flame by Asynchronous Optical Sampling", in *Process Diagnostics: Materials, Combustion, Fusion* (A. K. Hays, A. C. Eckbreth and G. A. Campbell, eds.), Materials Research Society, Pittsburgh, PA, 1988, pp. 187-192.

- B. J. Krielster, F. E. Lytto, G. J. Flechtner, Y. Jiang, G. B. King and N. M. Laurendeau, "Asynchronous Optical Sampling: A New Combustion Diagnostic for Potential Use in Turbulent, High-Pressure Flames", *Optics Letters*, March, 1989.
- G. J. Fiechtner, Y. Jiang, G. B. King N. M. Laurendeau, R. J. Kneisler and F. E. Lytle, "Determination of Relative Number Density and Decay Rate for Atomic Sodium in an Atmospheric Premixed Flame by Asynchronous Optical Sampling", *Twenty-Second Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, PA, in press.
- G. J. Fiechtner, G. B. King, N. M. Laurendeau, R. J. Kneisler and F. E. Lytle, "High UV Power from Synchronously Mode-Locked Dye Lasers: Dispelling the Myth", submitted to *Applied Spectroscopy*.

The progress of the ASOPS project during the past year has been presented at several meetings and seminars and has been well received by the scientific community:

- "Asynchronous Optical Sampling : A New Combustion Diagnostic for High-Pressure Flames," Pittsburgh Conference, New Orleans, LA, February 26, 1988.
- 2. "Asynchronous Optical Sampling: A New Generation of Picosecond Instrumentation," San Diego State University, San Diego, CA, March 7, 1988.
- "Asynchronous Optical Sampling : A New Generation of Picosecond Instrumentation," 1988 Francis Clifford Lecture, University of Pittsburgh, Pittsburgh, PA, April 13, 1988.

- "Determination of Relative Number Density and Quenching Rate for Sodium in an Atmospheric Premixed Flame by Asynchronous Optical Sampling," Spring Meeting, Central States Section/The Combustion Institute, Indianapolis, IN, May 1, 1988.
- "Async >nous Optical Sampling : A New Combustion Diagnostic for High-Pressure Flames," The Third Chemical Congress of North America, American Chemical Society, Toronto, Canada, June 5-10, 1988.
- "Determination of Relative Number Density and Decay Rate for Atomic Sodium in an Atmospheric Premixed Flame by Asynchronous Optical Sampling", Twenty-Second Symposium (International) on Combustion, Seattle, WA, August 15, 1988.
- 7. "Asynchronous Optical Sampling: A New Generation of Picosecond Pump/Probe Instrumentation," Eastern Analytical Symposium, New York, NY, October 3, 1988.

4. RESEARCH PERSONNEL

Professors Galen B. King and Normand M. Laurendeau in the School of Mechanical Engineering and Professor Fred E. Lytle in the Department of Chemistry are co-principal investigators for this research. Mr. Ronald Kneisler, a chemistry Ph.D. candidate, joined the group in January, 1986. Mr. Gregory Fiechtner, a mechanical engineering M.S. candidate, and Mr. Yan Li, a mechanical engineering Ph.D. candidate, joined the group in July, 1986 and January, 1988, respectively.

References

- 1. P. W. Fairchild, G. P. Smith and D. R. Crosley, J. Chem. Phys. 79, 1975 (1983).
- 2. R. P. Lucht, D. W. Sweeney and N. M. Laurendeau, Combust. Flame 50, 189 (1983).
- 3. N. S. Bergano, P. A. Jaanimagi, M. M. Salour and J. H. Bechtel, Opt. Lett. 8, 443 (1983).
- 4. R. Schwarzwald, P. Monkhouse and J. Wolfrum, Chem. Phys. Lett. 142, 15 (1987).
- 5. Y. Takubo, T. Okamoto, N. Komine and M. Yamamoto, Jap. J. Appl. Phys. 26, 416 (1987).
- 6. P. A. Elzinga, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, *Appl. Spectrosc.* **41**, 2 (1987).
- 7. P. A. Elzinga, R. J. Kneisler, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, *Appl. Opt.* **26**, 4303 (1987).
- 8. D. Stepowski and M. J. Cottereau, Appl. Opt. 18, 354 (1979).
- 9. D. Stepowski and M. J. Cottereau, Combust. Flame 40, 65 (1981).
- 10. T. Gray, Spectra Physics Corp., Mountain View, CA, personal communication (1987).
- 11. G. J. Fiechtner, Y. Jiang, G. B. King, N. M. Laurendeau, R. J. Kneisler and F. E. Lytle, *Twenty-Second Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, PA, in press.
- 12. R. A. Beaman, A. N. Davies, A. J. Langley, W. J. Jones, Chem. Phys. 101, 127 (1986).
- 13. Y. Takubo, T. Okamato and M. Yamamoto, Appl. Opt. 25, 740 (1986).
- 14. C. V. Shank, E. P. Ippen and Omar Teschke, Chem. Phys. Lett. 45, 291 (1977).
- 15. T. F. Johnston, Jr. and T. J. Johnston, *Laser Spectroscopy VI*, Springer Ser. Opt. Sci. **40**, pp. 417-418 (1983).
- 16. D. Welford, W. Sibbett and J. R. Taylor, *Opt. Comm.* **35**, 283 (1980).
- 17. N. Wang and U. Gaubatz, Appl. Phys. B 40, 47 (1986).
- 18. R. J. Kneisler, F. E. Lytle, G. J. Fiechtner, Y. Jiang, G. B. King and N. M. Laurendeau, *Opt. Lett.*, in press, March 1989.