
**1988 SPRING
TECHNICAL MEETING**



**CENTRAL STATES SECTION
The Combustion Institute**

**COMBUSTION FUNDAMENTALS
AND APPLICATIONS**

May 2-3, 1988

**Union Station Holiday Inn
Indianapolis, Indiana**

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MONDAY, MAY 2, 1988

Session B-1 COMBUSTION MEASUREMENTS I

Chairperson: George Lavoie, Ford Motor Co.

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DETERMINATION OF RELATIVE NUMBER DENSITY AND QUENCHING RATE FOR ATOMIC SODIUM IN AN ATMOSPHERIC PREMIXED FLAME BY ASYNCHRONOUS OPTICAL SAMPLING

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Introduction

Pump/probe methods are commonly employed to measure subnanosecond excited state processes in liquid and gas phase systems.^{1,2} Asynchronous Optical Sampling (ASOPS) is a newly-developed pump/probe method that will potentially allow the determination of number densities and relaxation rates in turbulent, high-pressure flames. In addition, ASOPS should yield a better signal-to-noise ratio than laser-induced fluorescence in practical combustion environments. The ASOPS method utilizes a coherent, signal-carrying beam and thus requires no more optical access than LDV measurements. The current ASOPS instrument consists of two dye lasers synchronously pumped by two frequency-doubled, mode-locked Nd:YAG lasers. The two lasers operate at slightly different repetition rates, causing a relative phase walk-out between the pump and probe beams. This strategy allows the mapping of subnanosecond excitation processes in a time scale on the order of milliseconds.

In previous papers,^{3,4} we reported initial ASOPS studies of Rhodamine B in methanol. We now extend the ASOPS technique to measurement of the relative number density of atomic sodium in an atmospheric CH₄/O₂/N₂ flame. The reconstruction of a fluorescence decay from the sodium D₂ transition (589.0 nm) is also used to determine the decay time of atomic sodium under flame conditions. To further demonstrate the viability of ASOPS as a combustion diagnostic, both ASOPS and laser-induced fluorescence (LIF) are used to measure relative concentration profiles and saturation curves. The results obtained using each technique are found to be in close agreement. Moreover, the signal-to-noise ratios of each method are of the same order-of-magnitude. We thus conclude that the ASOPS method has great potential for use in high-pressure, turbulent flames.

Operating Principles

In conventional pump/probe instruments the pump and probe lasers operate at identical repetition rates, and an optical delay line is used to control the relative timing between pulses from the two lasers. Moreover, some type of mechanical or electro-optical chopping scheme is generally employed to induce an amplitude modulation on the signal which facilitates the use of synchronous detection.^{5,6,7,8} In contrast, the ASOPS instrument employs pump and probe lasers operating at slightly different repetition rates. This induces a repetitive phase walk-out between the pump and probe pulse trains.

The ASOPS process is illustrated in Fig. 1a, which shows the excited state population produced by several pump pulses over which the temporal positions of several probe pulses have been superimposed. 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 immediately preceding pump pulse. This is equivalent to varying the optical delay in a conventional pump/probe instrument. The entire process of Fig. 1a 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 amplitude modulate either beam to employ synchronous detection.

Figure 1b illustrates the probe intensity which occurs owing to 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 $[(f_{\text{pump}}/(f_{\text{pump}}-f_{\text{probe}}))]$, where f is the repetition rate of the two lasers. The ASOPS technique is thus an optical analog of the sampling oscilloscope.

The beat frequency of the system is determined by the difference in laser repetition rates, i.e., $f_{\text{beat}} = f_{\text{pump}} - f_{\text{probe}}$. The inverse of the pump frequency, $1/f_{\text{pump}}$, gives the free temporal range of the ASOPS instrument, while the inverse of the beat frequency, $1/f_{\text{beat}}$, represents the collection time for a single decay. The total number of points, N , sampled during the decay is determined by $N = f_{\text{probe}}/f_{\text{beat}}$. 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, or $f_{\text{beat}}/(f_{\text{probe}} f_{\text{pump}})$.

Equipment And Procedure

Since the ASOPS technique requires that the pump and probe lasers operate at slightly different repetition rates, two independent, mode-locked laser systems are needed. Both the pump and probe beams are derived from Spectra-Physics model 375B dye lasers, which are synchronously pumped by frequency-doubled, mode-locked Spectra-Physics series 3000 Nd:YAG lasers. Rhodamine 6G is used as the laser dye. The mode-locking frequencies are generated by two Programmed Test Sources (PTS) model 160 frequency synthesizers operated in a master-slave configuration. Both the pump and probe beams consist of an ~82 MHz train of ~10 psec pulses, each tuned to 589.0 nm. The PTS synthesizers are accurate to 0.1 Hz; hence, the laser repetition rates are very stable. Average powers in excess of 250 mW are obtained using 800-900 mW of pumping power. The typical operating parameters for the present ASOPS system are summarized in Table I.

A block diagram of the instrument is given in Fig. 2. As shown, each beam is split as it emerges from the laser. The weaker fraction is used to generate a trigger signal at the beat frequency of the system. This is done by separately monitoring each beam with fast photodiodes (EG&G FND-100) and electronically mixing the outputs of the photodiodes with a double-balanced mixer (Anzac MD-141) operating in a high-speed gating mode.⁹ Further details are given by Elzinga et al.⁴ and Kneisler et al.¹⁰

The remaining fractions of both the pump and probe beams pass through polarization rotator-beam splitter combinations which provide a convenient means of varying the power in either beam. In addition, the vertical component of the pump beam is combined with the horizontal component of the probe beam at the second polarization beam splitter. The beams pass colinearly through a focusing lens, the flame, and a collimating lens. Finally, the beams are separated by a third polarization beam splitter, after which the probe beam is monitored by a single photodiode (EG&G SGD-100A). Great care is taken to ensure that the pump and probe beams are completely separated at the final beam splitter, so that only the probe beam reaches the photodiode. The colinear configuration was chosen to compensate for the relatively large amount of noise present in these first flame studies. Because of this configuration, the ASOPS signal is averaged over a pathlength of approximately 1 cm. In future experiments, spatial resolution will be enhanced by crossing the pump and probe beams.

The photodiode output is filtered with a 4LE50CD Texscan 50 MHz low pass filter to remove the individual 82 MHz pulses, and is then directed to the appropriate detection system. For the temporal studies, a Hewlett-Packard 54100A digitizing oscilloscope, triggered at the beat frequency of the system, is employed. Each point of the oscilloscope signal is the result of 2048 averages. For the nontemporal studies, which include the concentration profiles and saturation curves, a Stanford Research Systems model 510 lock-in amplifier is used to monitor a single Fourier frequency component of the ASOPS signal at the beat frequency of 10 kHz.

To provide a comparison with the ASOPS results, laser-induced fluorescence (LIF) was used to obtain additional concentration profiles and saturation curves. The output from one of the previously-described synchronously pumped dye lasers is amplitude modulated at 1 kHz using a PTi model OC 4000 chopper. The beam is then directed through a polarization rotator-beam splitter combination into the flame. Fluorescence is collected by a 15-cm focal-length lens at right angles to the laser beam. To insure consistency, the fluorescence collection volume includes the same 1-cm pathlength through the flame used for the ASOPS measurements. A specially wired RCA 1P28B photomultiplier¹¹ is used to detect the fluorescence. The photomultiplier signal is monitored using the lock-in amplifier at a reference frequency of 1 kHz.

An atomic absorption slot burner¹² is employed to support an atmospheric premixed CH₄/O₂/N₂ flame into which a sodium chloride solution is nebulized. The burner provided fairly constant conditions for about thirty minutes, after which it became less reliable. Atomic sodium was chosen because of the large absorption cross section of its 3S_{1/2}→3P_{3/2} electronic transition. Since the 589.0 nm wavelength of the transition lies near the peak of Rhodamine 6G tuning curve, ample laser power was available to perform the saturation measurements.

Results And Discussion

Figure 3 shows the ASOPS signal for atomic sodium; the signal compares favorably with that expected in Fig. 1. In this study, the pump and probe beam powers were set at 65 mW and 10 mW, respectively. The photodiode signal was amplified by a factor of 100. Based on a least squares fit, the equivalent decay time in is 8.9 μsec. Since the ASOPS technique performs a temporal transformation of the decay, the actual lifetime of the excited state under flame conditions becomes 1.1 nsec. This value is consistent with previous decay times (~1 nsec) reported in the literature.^{12,13}

The decay curve in Fig. 3 was obtained by averaging over 2048 separate single-decay curves. The peak signal-to-noise ratio of the average decay curve is 33. We believe that a considerable portion of this noise can be attributed to impedance mismatches at the trigger photodiodes and at the double balanced mixer. Corrections are currently being made in these devices to improve the signal-to-noise ratio. Furthermore, suitable upgrades are possible with respect to the performance of both the Nd:YAG laser systems and the detection electronics.

The ASOPS signal in Fig. 3 contains an additional downward feature at the end of its free temporal range that is not predicted by Fig. 1b. We believe that this additional shape is due to competition between the pump and probe lasers. Figure 1a assumes that each probe pulse does not perturb the ground state before the arrival of the next probe pulse. However, this will not always be true in the present experiments because both the pump and probe lasers excite the same transition. For example, the 8199th pump pulse will arrive only 3 psec after the 8198th probe pulse. Thus for this pump pulse to see an unperturbed population, the disturbance caused by the previous probe pulse would have to disappear in 3 psec. This condition is obviously impossible to satisfy.

To further demonstrate the viability of ASOPS as a combustion diagnostic, both ASOPS and LIF were used to obtain both relative concentration profiles and saturation curves. The horizontal ASOPS and LIF profiles are plotted in Fig. 4. The pump and probe beam powers are 120 mW and 80 mW, respectively. The 120 mW pump beam is also utilized for the LIF measurements. As shown in Fig. 4, the relative concentration profiles obtained using each method are nearly identical. Furthermore, the signal-to-noise ratio for ASOPS was of the same order-of-magnitude as that for LIF. The ASOPS method yielded an inverse relative standard deviation of 34, while the LIF method yielded a value of 44. As was stated above, we expect the ASOPS performance to improve significantly with suitable corrections in the instrument.

The saturation curves obtained using both ASOPS and LIF are plotted in Fig. 5. For the ASOPS method, both the pump and probe beams are again tuned to the sodium D₂ transition. The pump beam power is varied while the probe beam power is held constant at 80 mW. For the LIF method, the probe beam is blocked, while the pump beam power is varied in a similar fashion. Once again, the ASOPS measurements correspond closely with the LIF measurements.

Conclusions

The similarity in results obtained by the ASOPS technique and the LIF method demonstrates the applicability of ASOPS as a combustion diagnostic. This conclusion is significant since ASOPS has several useful advantages when compared to LIF. For instance, ASOPS does not require the typical collection optics of a fluorescence measurement as the ASOPS signal is contained entirely within the coherent probe beam. LIF studies also require knowledge of quenching rates that are often ambiguous or unavailable; the ASOPS technique inherently supplies such information. Although ASOPS and LIF yielded similar signal-to-noise ratios in the present study, we believe that major improvements in the current ASOPS instrument are possible.

Our ultimate goal is to use the ASOPS method to monitor molecular species such as the OH radical which occurs naturally in practical combustion environments. Thus

Our ultimate goal is to use the ASOPS method to monitor molecular species such as the OH radical which occurs naturally in practical combustion environments. Thus future experiments will explore the extension of ASOPS from the visible to the ultraviolet regime. This will require some minor modifications of the present instrument, such as the addition of frequency-doubling crystals.

Acknowledgments

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f_{pump}	81.5970000 MHz
f_{probe}	81.5870000 MHz
f_{beat}	10 kHz
Free Temporal Range	12.2 nsec
Collection Time (Single Decay)	100 μ sec
Samples per Decay	8200
Sampling Interval	1.5 psec

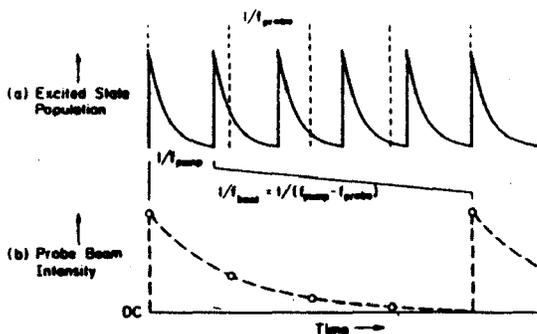


Figure 1. ASOPS timing diagram showing (a) excited-state population and (b) probe beam intensity. The vertical dashed lines in (a) represent probe pulses.

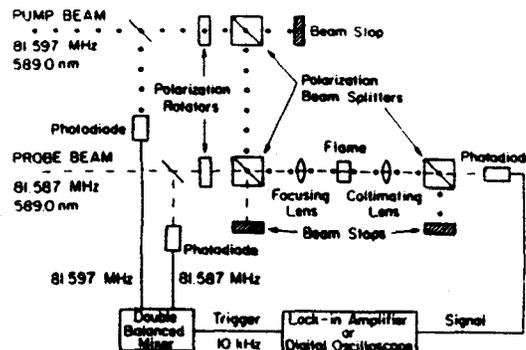


Figure 2. Block Diagram of the ASOPS instrument.

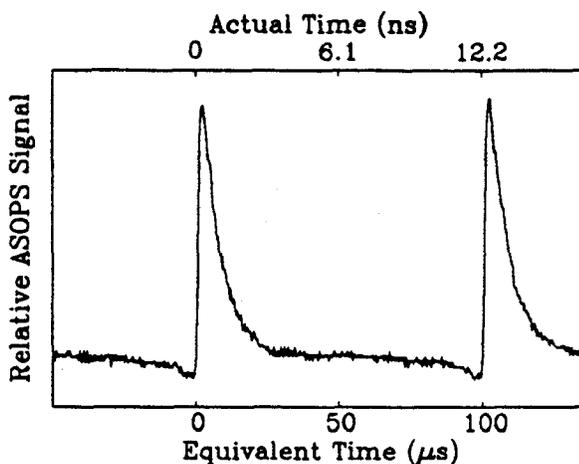


Figure 3. Temporal decay for atomic sodium obtained with both the pump and probe beams tuned to the D_2 transition (589.0 nm). The lower scale gives the equivalent time displayed on the oscilloscope, while the upper scale gives the actual time.

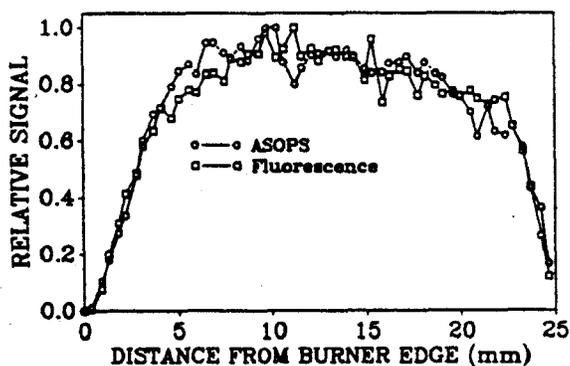


Figure 4. ASOPS and LIF horizontal relative concentration profiles. Measurements were taken -0.6 cm above the burner.

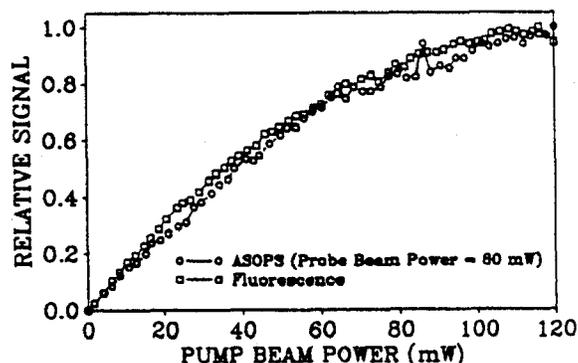


Figure 5. ASOPS and LIF pump-beam saturation curves. The ASOPS curve was obtained by varying the pump power, while the probe power was fixed at 80 mW.