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G. B. King, N. M. Laurendeau and F. E. Lytle

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This report describes the 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. Final results from the study of atomic sodium in an atmospheric flame are presented. The first ever UV pump/UV probe ASOPS signal for atomic indium is shown. Techniques for noise reduction are discussed along with initial results.



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

Progress Report
Air Force Office of Scientific Research
Grant No. AFOSR-89-0051
December 15, 1988 - December 14, 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 the 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. Final results from the study of atomic sodium in an atmospheric flame are presented. The first ever UV pump/UV probe ASOPS signal for atomic indium is shown. Techniques for noise reduction are discussed along with initial results.

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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 (SNR) 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 Progress to Date*

In the past year, we completed the ASOPS study of atomic sodium in an atmospheric flame. Attempts were made to obtain an ASOPS signal for the hydroxyl radical and no signal could be seen. Rhodamine B was again used as a sample to determine the cause of our difficulties but these studies were inconclusive. Therefore, we decided to use atomic indium as a sample since indium has a large Einstein coefficient for absorption, its transitions are in the UV and its concentration in the flame could be easily adjusted. Thus, we will first describe the sodium measurements and the UV measurements of atomic indium in a premixed flame. To eventually obtain the hydroxyl radical, steps must be taken to reduce the noise in the experiment. Noise sources in the experiment have been thoroughly classified and several noise-reduction remedies have been researched. Progress in noise reduction is shown with several frequency spectrum plots.

#### *2.1.1 ASOPS Study of Atomic Sodium*

The ASOPS detection of sodium in flames has been completed during the past year. In these experiments, a sodium chloride solution is seeded into a premixed flat $C_2H_4/O_2/N_2$ flame. The instrumental configuration of Fig. 1 is used, with the pump beam (20 mW) tuned to the $3S_{1/2} \rightarrow 3P_{3/2}$ transition (589.0 nm) and the probe beam (5 mW) tuned to the $3P_{1/2} \rightarrow 5S_{1/2}$ transition (615.4 nm). A peak SNR of 39:1 was obtained after 256 averages on the digitizing oscilloscope (Fig. 2).

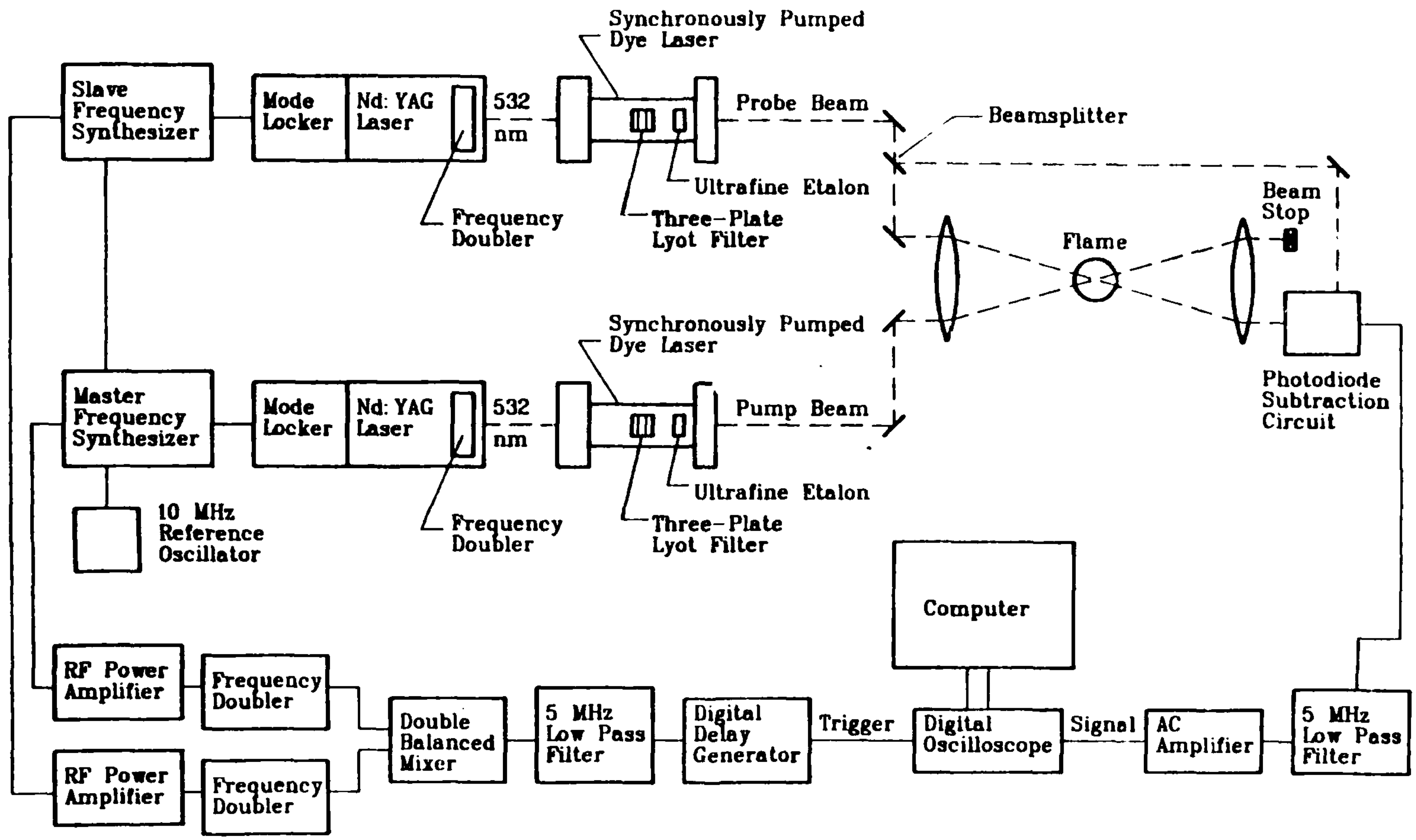


Figure 1. Diagram of the ASOPS instrument for the sodium studies.

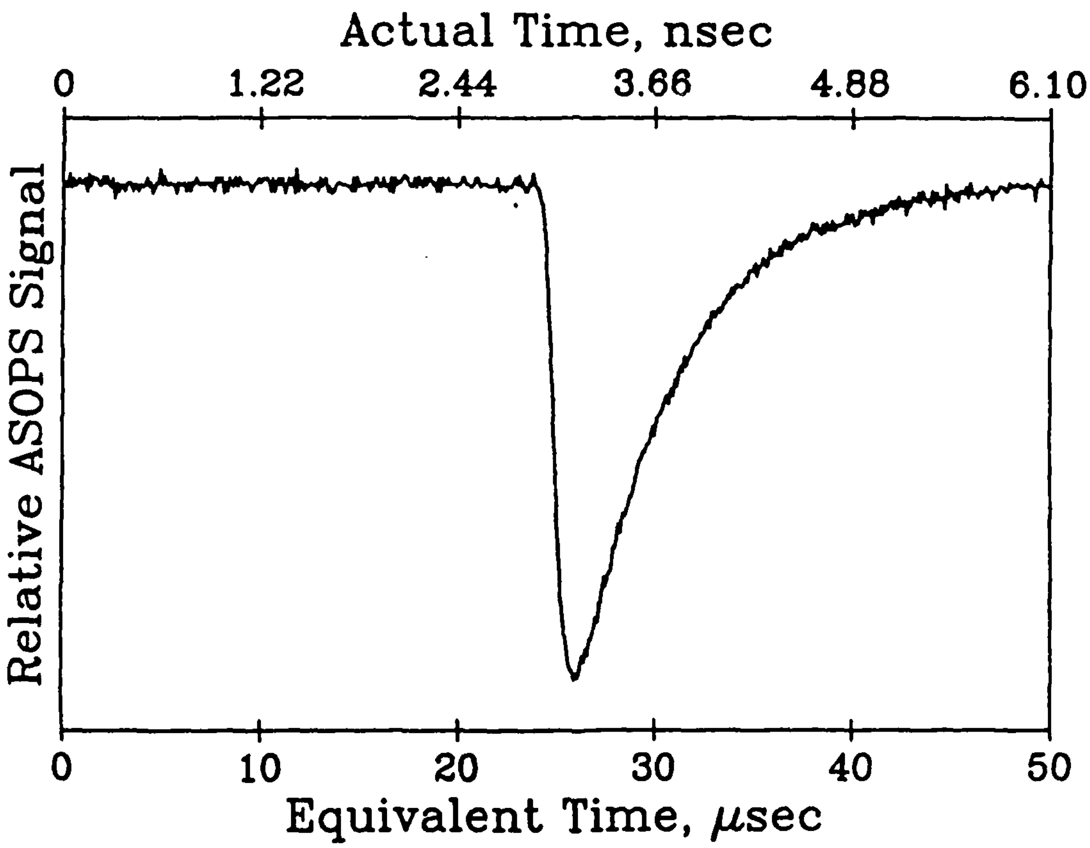


Figure 2. ASOPS signal for atomic sodium.

Temporal studies of the sodium results have also been completed. Here, a rate equation analysis is applied. The resulting curve fit to the data of Fig. 2 yields an electronic quenching rate of $Q=1.72\times10^9$ $sec^{-1}$. This value corresponds favorably with literature values ranging from $1.4\times10^9$ $sec^{-1}$ reported by Takubo et al[1] in a propane/air flame to $2.1\times10^9$ $sec^{-1}$ reported by Russo and Hieftje in a natural-gas/air flame.[2] Papers describing the results of the ASOPS sodium studies are presently being prepared for publication.

### *2.1.2 Initial UV Studies*

At the conclusion of the sodium studies, attempts were made to obtain an ASOPS signal for the hydroxyl radical and no signal could be seen. At that time it was uncertain whether the problem was lack of sensitivity or too much noise. Rhodamine B was again used as the sample and the following series of stepwise experiments were attempted. First, we obtained the ASOPS signal with both lasers in the visible. This verified that nothing had been drastically altered in the instrument. Second, the power of the pump laser was reduced to match the expected number of photons for a UV pump beam. Third, we obtained an ASOPS signal with a UV pump and a visible probe. This mitigated against any problems dealing with the introduction of the UV beams. Fourth, with both lasers in the visible, both the probe power and the pump power were reduced to again match the expected number of photons for a UV pump beam and a UV probe beam. At this point no signal could be seen. Therefore a new fluorophore was needed to determine the source of our problems. The new fluorophore chosen was indium.

### *2.1.3 Instrumental Developments*

There are a few changes in the instrument since the last report. A block diagram of the current ASOPS instrument is shown in Fig. 3. The pump dye laser contains only the three-plate Lyot filter while the probe dye laser contains both the three-plate Lyot filter and the ultra-fine etalon. The pump dye laser must be set up for maximum second-harmonic generation (SHG) while the probe dye laser needs to be as stable as possible and this configuration satisfies these criteria. The output of each dye laser passes through a focusing lens, a $LiIO_3$ crystal, a collimating lens and a Pellin-Broca prism. This provides the UV beams which then cross as they pass through the flame. The burner is a Perkin-Elmer atomic absorption burner with a specially designed Winefordner burner head. A $C_2H_2$/air mixture was used and an indium solution was aspirated into the flame. The photodiode subtraction circuit was used for the detector.

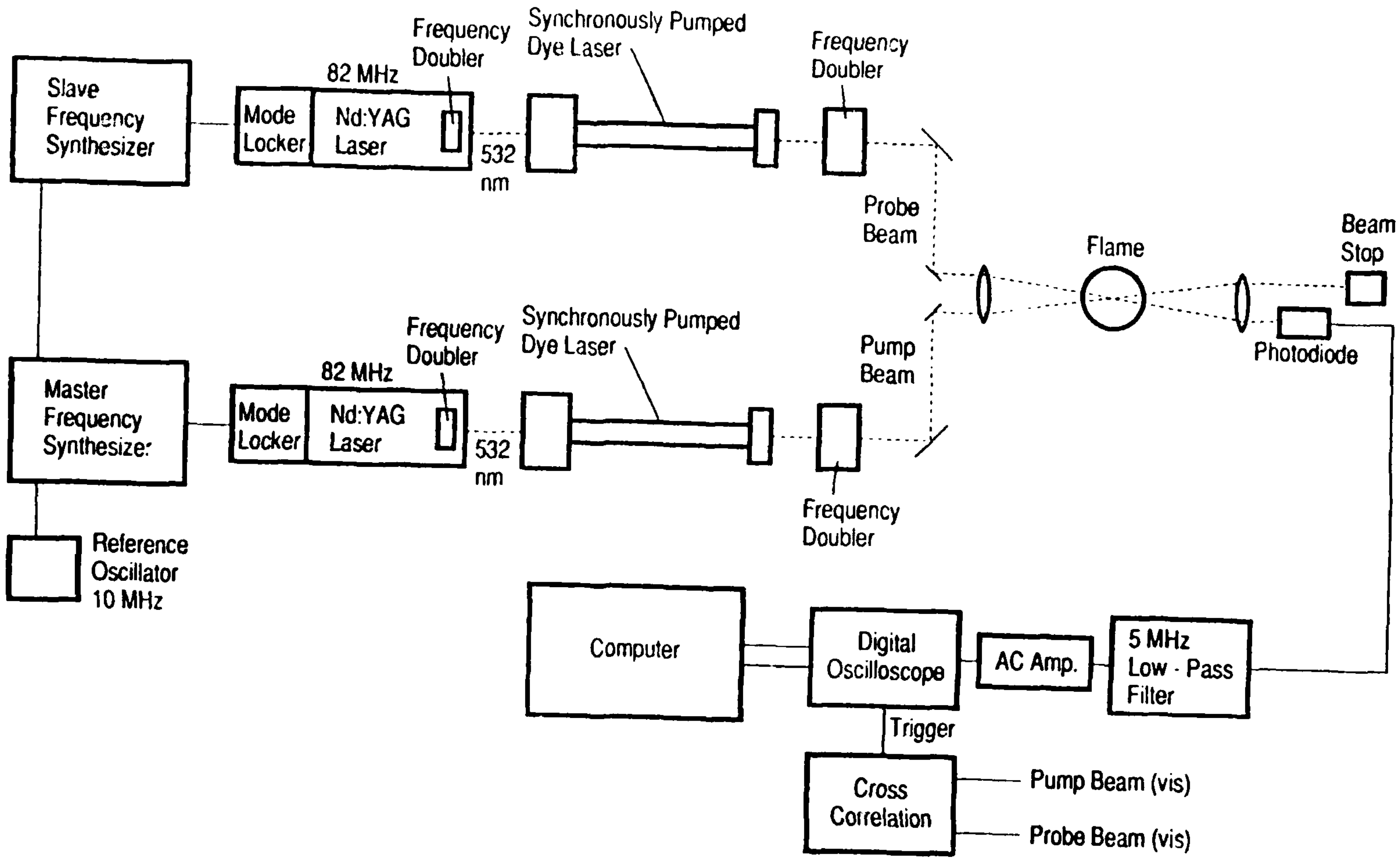


Figure 3. Diagram of the ASOPS instrument for the indium studies.

### *2.1.4 Experimental Results with Indium*

For the indium experiments, both the pump and probe beams were set to the same wavelength of 303.94 nm. The ASOPS signal is shown in Fig. 4. The pump power is ~ 20 mW while the probe power is ~ 1 mW. The Einstein coefficient for indium is approximately 1000 times that of OH. These results represent the first UV pump/UV probe signal obtained with the ASOPS methodology. Unfortunately, the spectroscopy of indium makes it almost impossible to use for modeling the ASOPS signal. Therefore, a new element such as magnesium will eventually be used.

### *2.1.5 Noise Reduction*

The next step in the research has been to reduce the noise in the experiment. The different trigger schemes used for ASOPS have had problems either with temporal jitter (optical photodiode mixing) or with feedback into the mode-locker drivers (electronic mode-locker driver mixing). We have returned to a slightly modified version of the optical cross-correlation trigger used previously.[3] The previous design used a colinear laser beam geometry which required the two wavelengths to be quite different. When the wavelengths were similar, a large background would result due to the second harmonic of the individual laser beams. In the new configuration, the two laser beams cross in the frequency doubling crystal. The generated second harmonic beam due to the cross correlation is then spatially separated from the remaining beams. This reduces the background the detector sees because an iris can be used to pass only the second harmonic due to the cross correlation. More importantly, this approach does not require the two lasers to be far apart in wavelength.

A Conoptics LASS-II laser noise eater has been tested with the probe laser. The detection system consisted of the photodiode subtraction circuit, a 50 ohm line driver and an AC amplifier. Fig. 5 shows the laser alone being detected by one of the photodiodes, the effect of using the subtraction circuit and the background level of the detector electronics. The latter trace is due to the output of the detection system with no laser present. The subtraction circuit reduces the laser background by a factor of 3 or more but the coherent spike at ~36 kHz is still quite substantial. In Fig. 6 the upper trace shows the effect of the noise eater only, and the bottom trace shows the combination of the noise eater and the subtraction circuit. The coherent noise spike at ~36 kHz has been reduced by a factor of 10 by the combination of the noise eater and the photodiode subtraction circuit as compared to the subtraction circuit alone

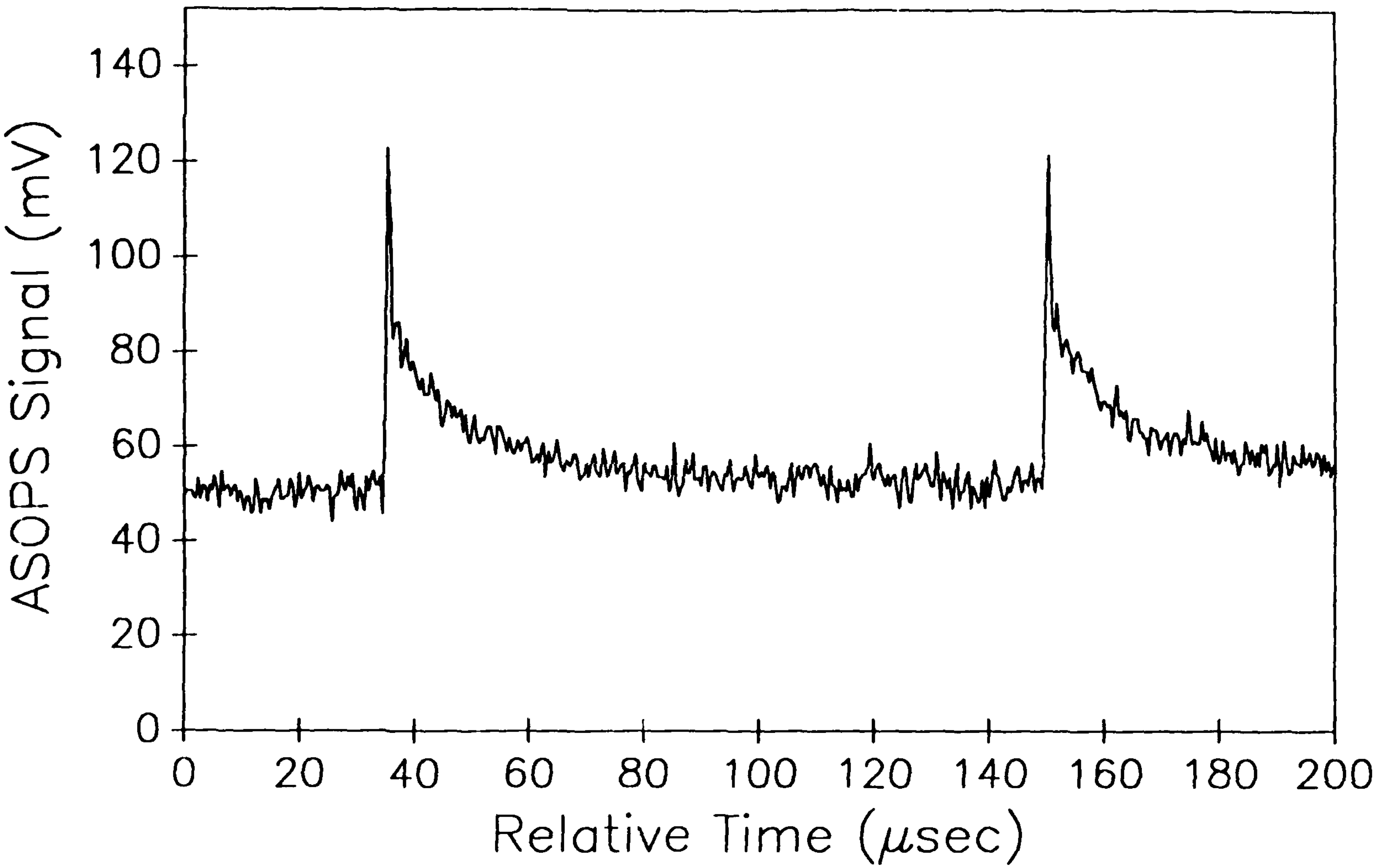


Figure 4. ASOPS signal for atomic indium.

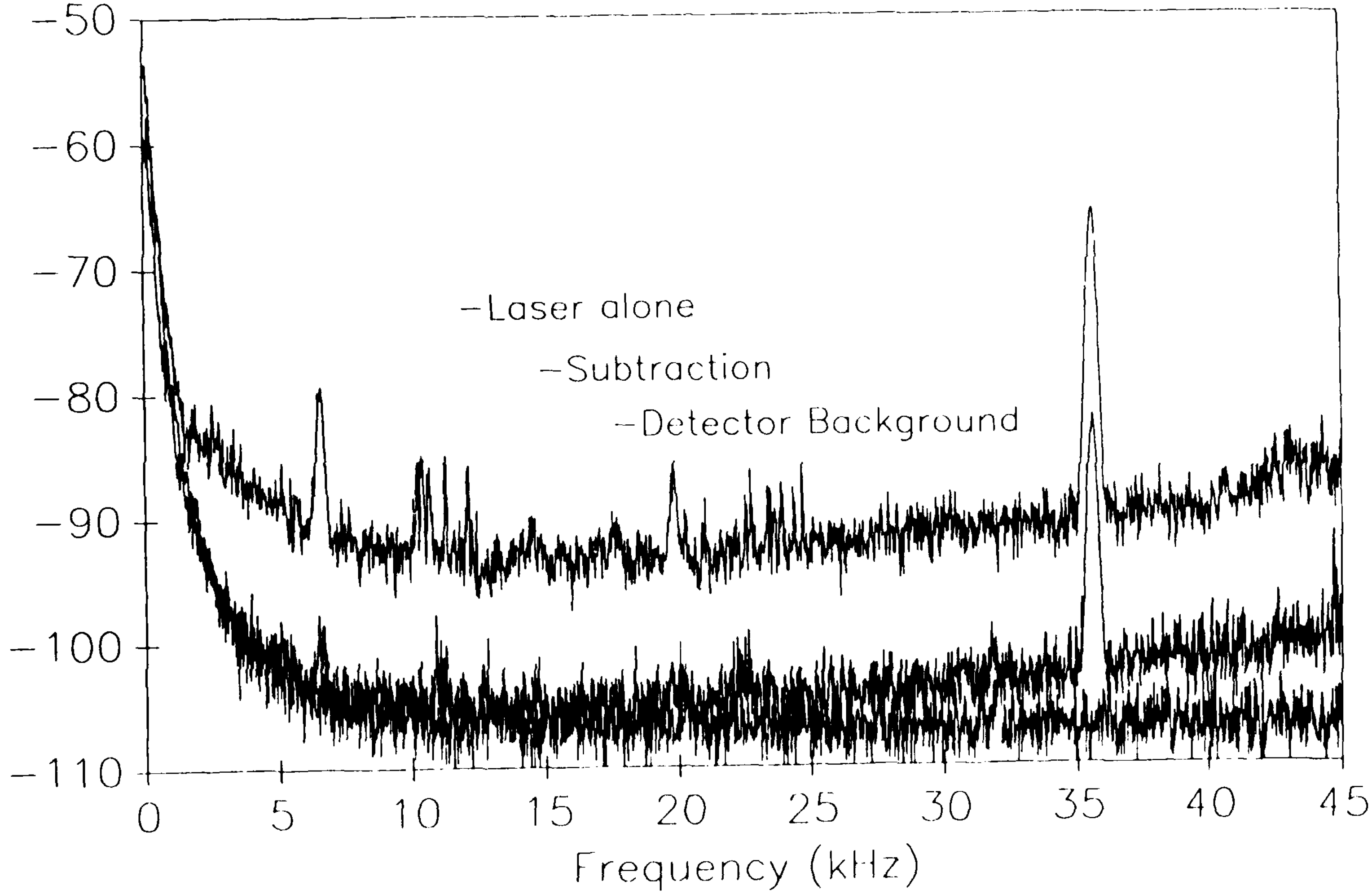


Figure 5. Frequency spectra of the laser alone, the laser with photodiode subtraction and the detection system background.

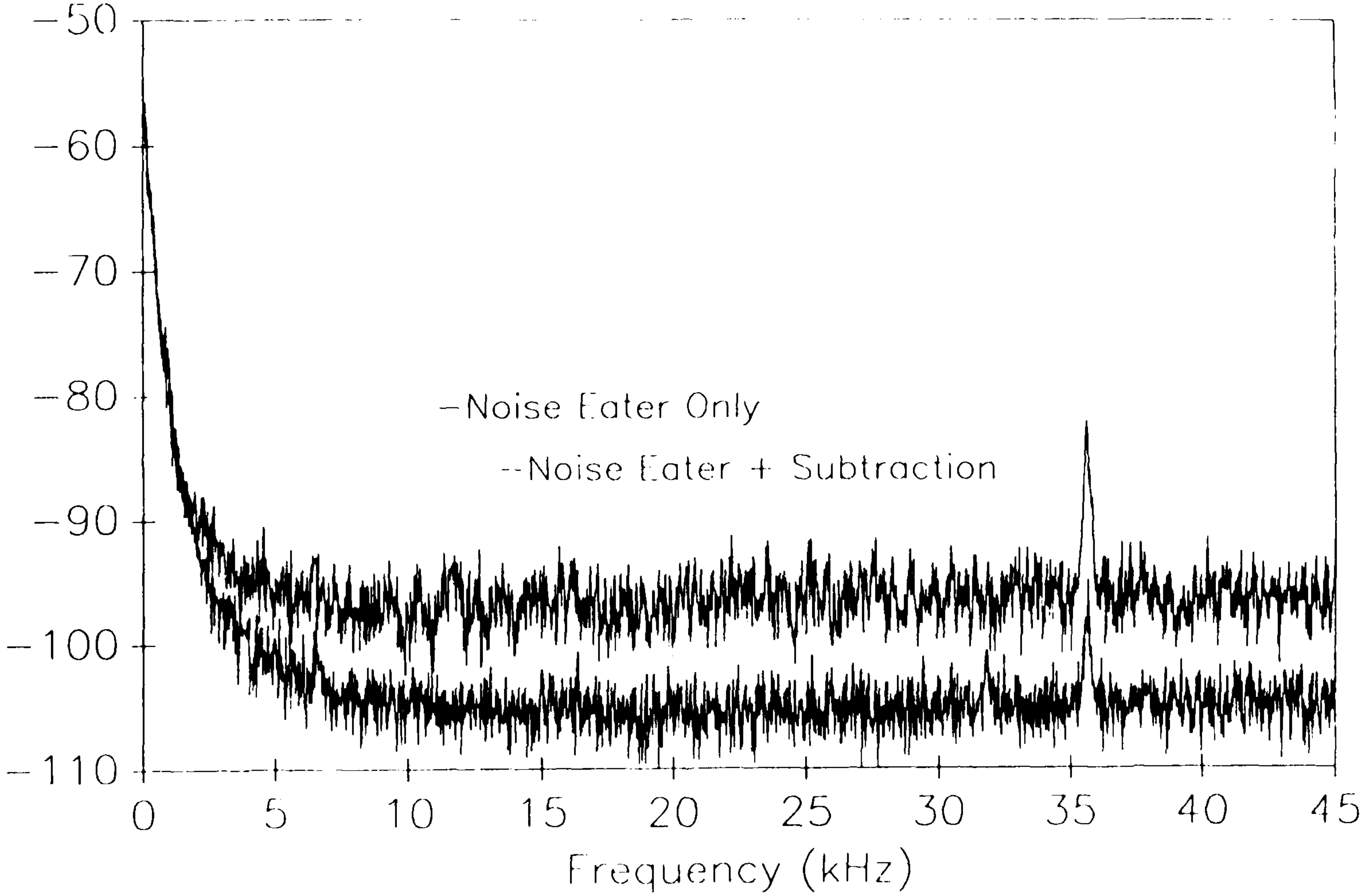


Figure 6. Frequency spectra of the laser with the noise eater and the laser with both the noise eater and photodiode subtraction.

and by a factor of 55 as compared to the laser alone. The combination of the two results in a background that is limited by the detection system. Therefore any improvements in the detection system electronics would benefit the ASOPS SNR.

### 2.2 Current Research

We are currently attempting to make ASOPS measurements of atomic magnesium because of its simple spectroscopy. The pump beam wavelength is 285.21 nm and the probe beam wavelength is 571.11 nm. The spectroscopy of magnesium allows for the use of a simple three-level ASOPS model. The concentration of magnesium will be varied to determine the lower limit of detection. With this quantitative information, we can directly determine expected ASOPS signals for OH. The other focus of the research is based on noise reduction.

As stated earlier, the combination of the noise eater and the photodiode subtraction circuit makes the ASOPS measurements detector limited. Dr. Robert Santini of the Instrument Design Group of the Department of Chemistry has looked at our photodiode subtraction circuit and believes he can improve the design to reduce the detector background. Our current circuit design only has a bandwidth of ~1 MHz which is not ideal. The new design will have an increased bandwidth between 5 and 10 MHz.

### 2.3 Future Work

The LASS-II noise eater has a bandwidth of 500 kHz. With a beat frequency of 10 kHz, Fourier components of the ASOPS signal out to ~ 5 MHz are desired. So we are currently waiting for an improved noise eater (LASS-III) with a bandwidth of 7 MHz. This should reduce the laser background even further in the frequency region of interest.

The laser power supplies are large sources of coherent noise due to their inherent switching circuits. Thus, multiple ~18 kHz frequencies appear directly on the laser beams. Work will be done on the power supply electronics to filter out this noise.

The dye laser also contributes to noise mainly from dye jet imperfections and poor cavity length control. New sapphire dye jets have shown a dramatic decrease in the noise bandwidth compared even to specially constructed steel jets.[4] New jet mounts will be machined for our dye laser to accommodate the sapphire jets. Piezoelectric output coupler cavity length controllers will be installed to give precise adjustment to counteract the effect of temperature variations.

Initially, we had decided to purchase a transient digitizer for our data collection system because it was the best equipment available. The digitizer is able to collect data rapidly but cannot average without skipping over incoming data thus increasing the time for data collection. Our view was to collect all the data in memory, then average in software rather than hardware after data collection was complete. In our conversations with Dr. David Bloom of the Department of Electrical Engineering at Stanford University, we have discovered that his group has looked at this very same data collection problem. Fortunately, his group was able to design a multi-channel boxcar averager that can average the data "on the fly". We are either going to purchase a completed version of this instrument or we may be able to obtain the schematics and have our instrument shop construct it. We are currently deliberating with the Bloom group as to the most reasonable approach.

## 3. PUBLICATIONS AND PRESENTATIONS

The following papers have been published.

1. 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, p.1915 (1988).
2. R. J. Kneisler, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, "Asynchronous Optical Sampling: A New Combustion Diagnostic for Potential Use in Turbulent, High-Pressure Flames," Opt. Lett. **14**, 260 (1989).
3. G. J. Fiechtner, G. B. King, N. M. Laurendeau, R. J. Kneisler and F. E. Lytle, "Efficient Frequency Doubling for Synchronously Mode-Locked Dye Lasers," Appl. Spectrosc. **43**, 1286 (1989).

The progress of the ASOPS project during the past year was presented at the following meeting.

1. "Measurement of Atomic Sodium in a Premixed Atmospheric Flame by Asynchronous Optical Sampling", Spring Meeting, Central States Section/The Combustion Institute, Dearborn, MI, May 2, 1989.

## 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 and should complete his Ph.D. thesis by the summer of 1990. Mr. Gregory Fiechtner, a mechanical engineering M.S. candidate, joined the group in July, 1986 and completed his M.S. thesis in August of 1989. He will continue on the ASOPS project as a Ph.D candidate. Mr. Brian Thompson has already committed to the ASOPS project, and will join us as an M.S. candidate in mechanical engineering in the summer of 1990.

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2. R. E. Russo and G. M. Hieftje, *Appl. Spectrosc.* **36**, 92 (1982).
3. P. A. Elzinga, R. J. Kneisler, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, *Appl. Opt.* **26**, 4303 (1987).
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