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of Atomic Potassium in Atmospheric
Flames Using a Regeneratively
Mode-Locked Ti:Sapphire Laser**

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QUANTITATIVE CONCENTRATION MEASUREMENTS OF ATOMIC POTASSIUM IN ATMOSPHERIC FLAMES USING A REGENERATIVELY MODE-LOCKED Ti:SAPPHIRE LASER

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Abstract

We demonstrate the use of mode-locked Ti:Sapphire lasers to make quantitative concentration measurements in turbulent, high-pressure flames. Turbulent combustion is an important factor in many practical energy conversion devices. Laser-induced fluorescence has been widely used to measure concentrations of flame radicals, even under conditions of turbulence. Unfortunately, laser-induced fluorescence measurements have been limited to probability distribution functions acquired at ~ 10 Hz because of the repetition rate of the lasers that are required. We seek to overcome this difficulty using picosecond pump/probe absorption spectroscopy.

Atomic potassium is seeded into an atmospheric, premixed CH_4/air flame. In all our measurements, the spatial resolution is 0.137 cm. We have also demonstrated the temporal resolution that the picosecond pump/probe instrument offers. Here, 50-Hz fluctuations were induced in an otherwise laminar flame using an audio speaker, and the resulting fluctuations in the potassium concentration were resolved on power spectral density (psd) plots. A detection limit of $1.5 \times 10^{11} \text{ cm}^{-3}$ was obtained for the psd system. A simple reduction in laser spectral bandwidth by a factor of 100 would reduce this detection limit to only $1.5 \times 10^8 \text{ cm}^{-3}$. We estimate that a corresponding detection limit of $3.6 \times 10^{12} \text{ cm}^{-3}$ for CH is possible. The temporal resolution of our system is presently limited to that of the borrowed detection electronics, and simple modifications will allow resolution beyond 1 MHz.

Introduction

In this paper we discuss the development and demonstration of an instrument for making spatially ($\sim 100 \mu\text{m}$) and temporally (less than $1 \mu\text{s}$) resolved concentration measurements of radical species in practical combustors. A wide variety of combustion

studies would benefit from this instrument. Existing laser diagnostic methods are not appropriate for measurements in many reactors. We demonstrate a new method for concentration measurements based on picosecond pump/probe absorption spectroscopy using regeneratively mode-locked Ti:Sapphire lasers. In this demonstration, atomic potassium is seeded into a premixed methane/air flame and the potassium concentration is then measured on a quantitative basis, calibrated with line-of-sight absorption spectroscopy. The flame is acoustically driven to simulate the fluctuating environment that is characteristic of practical combustors. The pump/probe signal is then manipulated using a fast-Fourier transform (FFT), and rapid fluctuations are thereby identified. The detection limit of the FFT system is measured. Based on these results, we recommend future experiments using this laser technology with the pump/probe method.

Motivation

The picosecond pump/probe absorption instrument that we demonstrate has a number of important features that should prove beneficial to combustion research:

1. Temporal resolution down to $\sim 1 \mu\text{sec}$ and spatial resolution down to $\sim 100 \mu\text{m}$.
2. Concentration measurements independent of the collisional environment.
3. Quantitative concentrations.
4. Detection of minor and major species, and species that do not have significant fluorescence yields.

We now consider some specific situations that would benefit from the pump/probe instrument.

Applications

Characterization of the fundamental properties of turbulent reacting flows is of critical importance. Models and experiments are necessary because both mixing and finite-rate chemical kinetics control the overall rate of reaction and the final product mix. It is therefore necessary to develop the capability

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to measure chemical species (major and minor) at the characteristic time and length scales in a turbulent flow field. Turbulence frequency spectra, which can often go out to 10 kHz, can be used to identify turbulence regimes and scales.¹ In the past, experimental work has centered on laser doppler anemometry and on corrected, fine-wire thermocouple measurements of velocity and temperature spectra; composition fields are likely to contain length and time scales different than those of the velocity field.²

It would therefore also be beneficial to make spatially- and temporally-resolved statistical concentration measurements of key species in turbulent flames. Turbulence length and time scales cannot be resolved if the measurement scale or time is greater than the smallest (Kolmogorov) scales of interest, which are typically hundreds of microns in length and less than one hundred microseconds in time.³ A measured concentration power spectral density (psd) can be used to infer all the length and time scales of importance. Moreover, a system which is capable of detecting psd's is also capable of acquiring probability density functions (pdf's), but at a much faster data collection rate.

Another example is ignition, which can require measurements to be completed on a millisecond time scale.^{7,8} The event may take place at a known, periodic rate, such as ignition in an internal combustion engine, or it may occur at an unpredictable instant in time, as with the study of ignition of bulk metals.^{9,10} Our instrument may even be able to probe concentrations during ignition in detail, giving information such as psd's and pdf's during the transient event.

Existing Diagnostics

Optical methods of measuring minor species concentrations have provided new insights into the combustion process.¹¹ Although a variety of techniques has been successfully applied to flames, such as absorption and laser-induced fluorescence (LIF), these techniques have major drawbacks for use in practical, high-pressure, fluctuating combustion environments. The most widely used laser-based diagnostic is LIF,¹² because its favorable signal-to-noise ratio (SNR) allows detection of minor species such as the hydroxyl radical. In turbulent flames, fluctuations in the collisional environment can cause uncertainty in the LIF signal.^{12,13} Corrections for this uncertainty have been made by simultaneous Rayleigh scattering measurements.¹⁴ However, conventional Q-switched laser technology is necessary to obtain sufficient photons for practical measurements. These lasers exhibit repetition rates of ~100 Hz, requiring single-shot measurements of probability distribution functions. Even with successful single-shot results, information between pulses is lost. In many cases, single-shot data are not possible due to inadequate signal-to-noise ratio (SNR), with ~10 averages necessary. Not all species have significant fluorescence yields, so LIF is appropriate for a limited set of molecules. Although LIF will always be applicable to many combustion studies because of its utility, there remain a number of cases where it is not adequate.

Excellent temporal resolution has been demonstrated for laser absorption measurements.¹⁵ When the signal is generated by a cw laser, the speed of the measurement is limited by the electronics used. All species absorb radiation, so this approach can be applied to a very wide range of molecules. Unfortunately, absorption is a line-of-sight technique, and thus has poor spatial resolution.

Coherent Anti-Stokes Raman Spectroscopy (CARS) is a two-photon nonlinear technique, so it has much lower sensitivity than LIF. It is a background-free four-wave mixing process. CARS produces a coherent, collimated and spectrally bright signal beam. This makes it possible to spatially filter spurious scattered light from the signal beam, and it requires less optical access than LIF. CARS is also relatively insensitive to pressure. Analysis of the CARS signal, however, requires complex spectral modeling. Because it is a nonlinear process with small cross sections, CARS requires the use of high power pulsed lasers, limiting temporal resolution. Because of the low sensitivity, it is typically applied only to major species like nitrogen.

An example of a spatially and temporally resolved species psd measurement is presented by Wrobel and Pratt,¹⁶ who used a cw dye laser to observe sodium fluorescence in a turbulent flame. The resulting signals were fast-Fourier transformed out to 10 kHz. Their same instrument could be used to acquire pdf's. While pdf's can be measured using pulsed lasers, cw diagnostics increase the data rate significantly. Wrobel and Pratt¹⁶ encountered systematic problems related to the use of low power dye lasers, and they quote $\pm 30\%$ uncertainty in the sodium measurement due to collisional quenching of fluorescence. While their work was a significant step, we use both a new laser device and new diagnostics.

Picosecond Pump/Probe Absorption Spectroscopy.

To overcome the difficulties associated with existing laser diagnostics, we have pursued an instrument based on the conventional pump/probe configuration.¹⁷ A typical example is shown in Figure 1. Here, the output of a picosecond laser system is split into two beams, the pump and the probe, so that both beams have the same pulse repetition rate. The pump is modulated with a chopper for low frequencies (<10 kHz), and an electrooptic modulator for higher frequencies (>1 MHz). The pump and probe are crossed in the flame using an LDV configuration, and the pump beam is directed to a beam stop after the flame. The probe is detected after the flame using either a photodiode or a photomultiplier. The signal is sent to a lock-in amplifier, which is triggered by the modulator frequency. An optical delay line is placed in the pump-beam path before the flame as a convenient way to temporally overlap the pump and probe pulses in the flame where the two beams cross. Because gasdynamic events occur on >100 ps time scales, picosecond pulses are used to remove any dependence on the collisional environment.

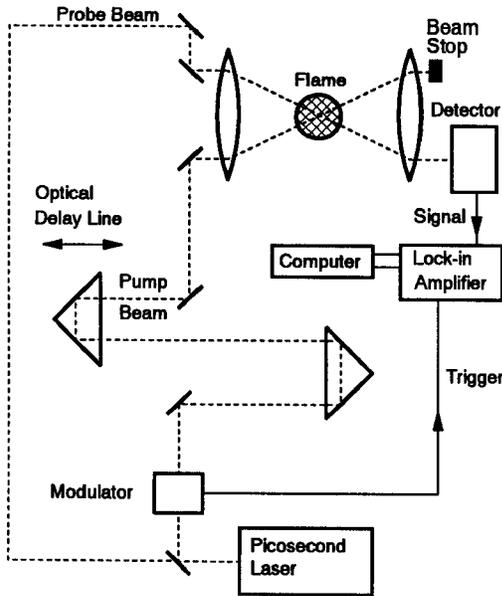


Figure 1. Conventional pump/probe instrument.

The pump/probe instrument has been extensively applied in the past. Initial demonstrations of the pump/probe method in a flame environment used a synchronously mode-locked dye laser as the source of picosecond pulses.¹⁸ The pump was modulated at megahertz frequencies, and the $2S_{1/2}-2P_{1/2,3/2}$ transition (671 nm) of atomic lithium, which had been seeded into the slot burner, was detected. An impressive detection limit $6.5 \times 10^4 \text{ cm}^{-3}$ was obtained for a lock-in time constant of 1 second; this detection limit is corresponds approximately to the limit defined by shot noise on the probe laser. Although the scope of these initial demonstrations did not include minor species diagnostics in flames, Fiechtner *et al.*¹⁹ applied a rate equation analysis to estimate that a corresponding detection limit for OH would be $2.3 \times 10^{12} \text{ cm}^{-3}$ for synchronously mode-locked dye lasers. Unfortunately, synchronously mode-locked dye lasers were found by Fiechtner *et al.*¹⁹ to lack sufficient power for detection of important minor species such as OH, CH, and NO. In the present study, we seek to retain the potential of the previous tests by replacing the synchronously mode-locked dye lasers with regeneratively mode-locked Ti:Sapphire lasers. Ti:Sapphire lasers have a number of advantages, including an improvement in average power by a factor of up to 1000 over older synchronously mode-locked dye lasers. This should allow an improved detection limit for species such as CH, as discussed below.

In our experiments, the pump and probe beams are crossed in the flame with the geometry of Figure 2, and both beams are in resonance with the same transition. The molecules will change the irradiance of the probe beam where it intersects the pump. This change in irradiance is defined as the modulation depth. In the present case, the modulation depth can be expressed^{19,21}

$$\alpha_{\text{MOD}} = 0.777 \frac{g_2}{g_1} \left(1 + \frac{g_2}{g_1}\right) \frac{c^4 A_{21}^2 P_{\text{AVE}}^{\text{PUMP}} N_{\text{T}} L}{16\pi^3 D^2 h \nu_{12}^5 f^L (\Delta\nu_{1/2}^L)^2}, \quad (1)$$

where h is Planck's constant, c is the speed of light, and the remaining variables and their present values are given in Table I. The pump/probe signal is proportional to the modulation depth multiplied by a detector efficiency, multiplied by the probe irradiance. Calibration by line-of-sight absorption yields the detector efficiency, placing results on a quantitative basis.

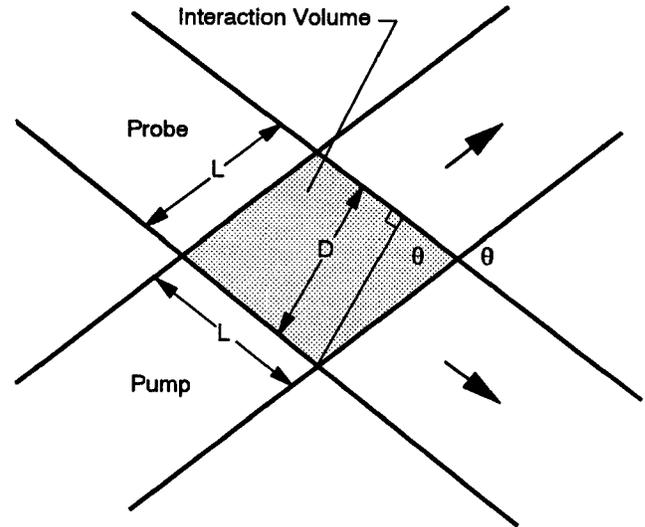


Figure 2. Crossed-beam geometry used in our experiments.

Table I. Operating parameters and important constants for present experiments. Variables are used in Equation (1).

SYMBOL	DESCRIPTION	VALUE
g_1	ground state degeneracy	2
g_2	excited state degeneracy	4
D	focal diameter	116 μm
L	interaction length	0.137 cm,
f^L	laser repetition rate	82 MHz
ν_{12}	1-2 transition frequency	$3.912 \times 10^{14} \text{ s}^{-1}$
$\Delta\nu_{1/2}^L$	laser bandwidth, FWHM	$2.29 \times 10^{11} \text{ s}^{-1}$
$P_{\text{AVE}}^{\text{PUMP}}$	average pump power	$4.4 \times 10^{-3} \text{ W}$
A_{21}	Einstein coefficient for spontaneous emission	$0.4 \times 10^8 \text{ s}^{-1}$

Choice of a Picosecond Laser

Mode-locking is one of several techniques for producing short pulses from lasers. We prefer this approach because the laser is cw pumped (vs. pulsed), providing cw output and cw data rates (40 - 50 MHz). Moreover, mode-locking enforces excellent longitudinal mode stability. There are numerous, compelling advantages associated with the use of such a laser :

1. Mode-locked lasers have high (> 80 MHz) repetition rates, so it is possible to achieve excellent signal-to-noise ratios with wide bandwidth using phase sensitive detection.
2. Mode-locked laser diagnostics are capable of recovering all the relevant frequency spectrum information, such as pdf's and psd's.
3. Mode-locked lasers also allow continuous monitoring of rapid events, such as ignition.
4. Mode-locking gives excellent longitudinal mode stability, eliminating the pulse-to-pulse mode hopping experienced with multi-mode pulsed laser systems.
5. With efficient mode-locking, pulses are transform-limited. Thus a measurement of temporal pulsewidth (easily accomplished) yields a reliable estimate of the spectral bandwidth.
6. Mode-locked lasers provide a beam with a well-defined TEM-00 spatial profile characteristic of many cw lasers. This allows us to obtain a good estimate of the spatial resolution of the instrument (the interaction length).
7. Perhaps most importantly, mode-locked lasers enable a minimum detectable modulation depth of 10^{-8} , corresponding to the shot noise limit.²⁰

In contrast, signals generated by pulsed lasers usually require averaging (often over 20 or more pulses), because single shot measurements are accompanied by large variability. The turbulence frequency spectrum that can be resolved is limited to half of the rate at which the laser is pulsed (10 Hz to 500 Hz), divided by the number of averages required; this restricts the type of flowfield that can be studied. Moreover, pulsed, single-shot measurements utilize much larger peak power. They become limited by saturation or breakdown of the medium, and the detector dynamic range must be large.

Apparatus and Procedure

Our complete experimental layout is similar to that of Figure 1. We use a Spectra-Physics regeneratively mode locked Ti:sapphire laser, equipped with a 2 ps Gires-Tournois interferometer.²² This laser produces about 900 mW of output when pumped with 7 W from an Ar:Ion laser, with autocorrelation pulse-widths of 1.4 ps (assuming a sech² pulse shape). In the present studies, the output from the Ti:sapphire laser is split into two portions: the pump beam is directed through a 1-kHz chopper, and is then crossed

with the probe beam in the flame with an inclusive angle of 4.85°. The pump-beam modulation is then impressed on the population being measured, which in turn modulates the probe beam in the interaction volume. For our geometric configuration, the interaction length is 0.137 cm. After the flame, the pump beam reaches a beam stop, and the probe beam is detected using a RCA 1P28 photomultiplier tube. The photomultiplier current is synchronously demodulated using a lock-in amplifier. The probe pulses are delayed by approximately 2 pulsewidths relative to the pump pulses to prevent the influence of a coherent spike on the pump/probe absorption signal.^{19,21}

The burner is a Perkin-Elmer aspirating unit fitted with a Meeker type burner head, operated with an equivalence ratio of 0.92 for the CH₄/air flame. Various solutions of KCl in water are aspirated into the flow in order to provide controlled, reproducible levels of atomic potassium in the flame. We perform these experiments with potassium because it has strong lines near the peak of Ti:sapphire. In the pump/probe experiments described below, the laser is tuned to the $4^2S_{1/2}-4^2P^o_{3/2}$ transition of atomic potassium at 766.5 nm. To avoid optical saturation of this transition, the pump power is set to 4 mW and the probe power is set to 5 mW.^{19,21} All concentrations of atomic potassium are optically thin.

A line-of-sight absorption measurement is performed for calibration with our pump/probe data. Here, several different solutions, with different concentrations of KCl, are atomized into the flame. Light from a tungsten lamp is passed through the flame and then detected with an optical multichannel analyzer (OMA); the OMA signal is integrated for 26.7 μ sec and averaged 4095 times. The absorption of the lamp emission by the $4^2S_{1/2}-4^2P^o_{3/2}$ transition of atomic potassium is measured and used to calculate the potassium concentration in the flame. Confidence intervals (95%) result from the uncertainty in calculating the integrated lamp absorption, due largely to OMA detector noise. The same KCl solutions are then atomized into the flame during picosecond pump/probe measurements, for which a lock-in bandwidth of 1 Hz is used. For each concentration, 6 pump/probe measurements are taken, and the spread of values is used to estimate 95% confidence intervals.

Finally, to demonstrate that this system can detect rapid concentration fluctuations, a small audio speaker on the fuel/air line is driven with a frequency synthesizer at 50 Hz. The loud speaker induces a known disturbance in an otherwise laminar flame. The visible flat flame front then oscillates vertically above the burner surface through a distance of approximately 1/2 cm. Stronger fluctuations can be produced by increasing the voltage to the speaker, but these disturbance levels often extinguish the flame. Despite the large spatial fluctuation of the reaction zone position, no significant beam steering is observed. Since the pump beam is modulated at 1 kHz, the flame disturbance also results in 50-Hz sidebands about the 1-kHz signal. The lock-in amplifier (an Ithaco model 395) bandwidth is set to its maximum of 200 Hz, so that the 50-Hz frequency

remains on the lock-in output.²³ Our choice to study a 50-Hz flame disturbance in no way implies a bandwidth limitation to the picosecond pump/probe instrument. In fact, using commercially-available equipment, the instrument has the potential to measure fluctuations over 10 MHz. The present frequency limit is defined by the existing lock-in amplifier. The synchronously demodulated lock-in output is directed to a PC based A/D system with an FFT board. The signal is sampled in record lengths of 8192 points at a rate of 500 Hz. This record length is then divided into 8 individual bins of 1024 points, which are FFT'ed and averaged together. This is repeated for all KCl solutions, allowing an estimate of the FFT-system detection limit.

Results and Discussion

The picosecond pump/probe absorption signal was recorded for several different potassium concentrations. Simultaneous line-of-sight absorption measurements were recorded, and pump/probe results were thereby made quantitative. The temporal resolution of our instrument is demonstrated in Figure 3. The FFT was obtained for a potassium concentration of $6.3 \times 10^{11} \pm 7 \times 10^{10} \text{ cm}^{-3}$, and shows the 50 Hz modulation. A subharmonic flame fluctuation at 25 Hz is also observed, emanating somewhere in the burner system, although we make no attempt to explain a mechanism for its presence. When the potassium concentration is varied, and the peak SNR of the 50-Hz spike is recorded, the detection limit corresponding to a SNR of unity is found to occur at $1.5 \times 10^{11} \pm 7 \times 10^{10} \text{ cm}^{-3}$. The

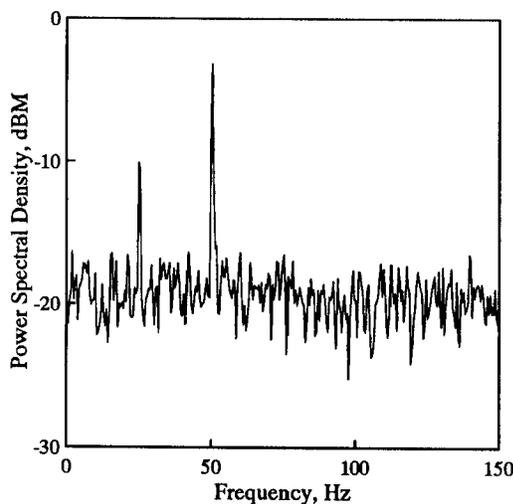


Figure 3. Demonstration of the temporal resolution of our instrument. The FFT of the lock-in amplifier output is obtained at a sampling rate of 500 Hz, and represents the average of 8 individual FFT's. The flame is driven with an audio speaker at 50 Hz. The FFT is obtained for a potassium concentration of $6.3 \times 10^{11} \text{ cm}^{-3}$ and clearly shows the 50 Hz modulation. A subharmonic flame fluctuation at 25 Hz is also observed.

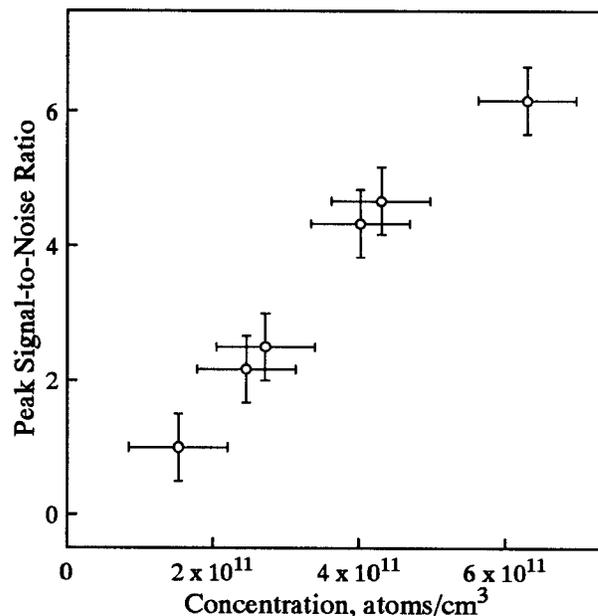


Figure 4. Plot of the peak SNR of the 50-Hz spike, shown in Figure 3, vs. potassium concentration. As expected, the SNR is linear with concentration. The detection limit, defined for a SNR of unity, is obtained for a concentration of $1.5 \times 10^{11} \text{ cm}^{-3}$.

remaining peak SNR's are shown in the main portion of Figure 4. As expected, a plot of SNR versus concentration is linear. The noise we encountered was caused by the lack of an instrumentation amplifier, and as a result, the signals were of a magnitude close to noise in the A/D converter. Amplification of the lock-in output would therefore directly increase the SNR of the FFT system. The modulation rates (mechanical chopper and acoustic loud speaker) were all slow because we were using borrowed equipment, none of which is fast. Lock-in amplifiers with higher bandwidths (10 kHz) are now commercially available. If necessary, a demodulator could be constructed to provide even wider bandwidths.²³

Future measurements will be done on flame radicals. This will require that the Ti:Sapphire laser radiation be frequency doubled or mixed to produce the desired wavelength. We are presently adding the necessary nonlinear crystals. For previous pump/probe measurements with synchronously mode-locked dye lasers, the detection of OH was not possible because of the notoriously low average power that the laser systems produced in the UV.^{19,21} We are much more optimistic for the present instrument. Nebel and Beigang²⁴ have recently demonstrated that second, third, and fourth harmonic generation from a mode-locked Ti:Sapphire laser allows tunable radiation from 205 nm to 525 nm. Maximum output powers of 700 mW, 120 mW, and 10 mW were obtained at wavelengths of 410 nm, 272 nm, and 210 nm, respectively. For each case an 82-MHz train of ~ 1.5 -ps pulses was studied. This new laser technology thus allows at least two orders of

magnitude more UV power than for previous laser systems; moreover, this additional laser radiation need not be attenuated, since the predicted saturation irradiance for diatomic species such as OH is ~ 900 mW.^{19,21} In addition, we note that several diatomic species fall within the frequency-doubled Ti:Sapphire laser tuning range, including CH (${}^2\Delta-{}^2\Pi$) at 431.5 nm and CN (${}^2\Sigma-{}^2\Sigma$) at 400.0 nm. Using the pump/probe model of Fiechtner *et al.*^{19,21} for CH with a pump power of 700 mW, we obtain a detection limit for the FFT system of 3.6×10^{16} cm⁻³. The present laser bandwidth is ~ 100 times too broad for efficient resonant interaction with gas phase species. This results in a large amount of background light that is not modulated but contributes to noise. A reduction in bandwidth by a factor of 100 would improve the SNR by 10,000 in accordance with Eq. (1). For the present Ti:Sapphire laser systems, such an improvement in bandwidth can be accomplished by changing the Gires-Tournois interferometer²² and installing a three-plate birefringent tuning filter. The corresponding detection limit for potassium would then be 1.5×10^7 cm⁻³, and for CH the resulting detection limit is estimated to be 3.6×10^{12} cm⁻³.

Previous studies with synchronously mode-locked dye lasers were hampered by sizable baseband noise on the laser output that was amplified by the frequency doubling process.²⁵ Although we are not aware of any noise comparisons between Ti:Sapphire lasers and mode-locked Nd:YAG lasers, Kafka *et al.*²² found the noise on the former to be favorable, while Son *et al.*²⁶ found that the noise from a mode-locked Ti:Sapphire laser is favorable when compared to that from a colliding-pulse mode-locked dye laser. Because of the improved output power and low noise from this new technology, we believe that picosecond pump/probe absorption spectroscopy could eventually become an important diagnostic in turbulent, high-pressure flames.

Summary and Conclusions

We have successfully demonstrated an instrument based on picosecond pump/probe absorption spectroscopy for measuring absolute concentrations in a rapidly fluctuating flame environment at practical operating pressures. In all our measurements, the spatial resolution was 0.137 cm. We have also demonstrated the excellent temporal resolution that the picosecond pump/probe instrument offers. Here, 50 Hz fluctuations in the potassium concentration were easily resolved on power spectral density plots. A detection limit of 1.5×10^{11} cm⁻³ was obtained for the psd system. Using the picosecond pump/probe absorption model, we find a corresponding detection limit of 3.6×10^{16} cm⁻³ for CH; for an improvement in bandwidth-coupling by a factor of 100, the detection limit improves to 3.6×10^{12} cm⁻³, of potential value to the combustion community. The temporal resolution of our system is presently limited to that of the borrowed detection electronics, and based on the success of these experiments, we are also obtaining electronic equipment to extend our resolution beyond 1 MHz.

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