Picosecond degenerate four-wave mixing on potassium in a methane–air flame

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We demonstrate that picosecond mode-locked laser-based degenerate four-wave mixing can be detected with good signal-to-noise ratios in an optically thin flame and that detailed turbulence statistics can be acquired by use of this technique. A regeneratively mode-locked Ti:sapphire laser was tuned to the $4^2S_{1/2}-4^2P^\circ_{1/2}$ transition in atomic potassium (which was doped into the flame) at 769.9 nm. Using the all-forward degenerate four-wave mixing geometry, we achieved signal-to-noise ratios of 70:1 without the use of a spatial filter. A sensitivity curve and a method for acquiring turbulence statistics are presented.

The need for quantitative measurement of minor species concentration and temperature in flames has lead to a number of recent studies based on resonant degenerate four-wave mixing[1] (DFWM). Unfortunately recent nanosecond pulsed laser studies have shown that the formation of thermal gratings can make it difficult to relate the observed signal to the number density.[2] Rakestraw *et al.*[3] pointed out that the use of pulses shorter than 100 ps can be used to avoid thermal gratings because generation of a thermal grating takes much longer. Conversely, an 82-MHz stream of picosecond pulses could be used to investigate thermal-grating formation and removal mechanisms in detail.

A measurement of the power spectral density of species concentrations in a turbulent flame could be used to identify important turbulence regimes and scales. Full turbulence spectra cannot be resolved, however, if the measurement scale or time is greater than the smallest (Kolmogorov) scales of interest, which are typically hundreds of micrometers in length and less than 100 μs in time (>10 kHz). The turbulence frequency that can be resolved by a high-power nanosecond pulsed system has an upper limit set by half the laser pulse repetition rate (10 Hz to as high as 500 Hz). In contrast, cw mode-locked lasers (emitting a stream of short pulses at 70–100 MHz) are capable of recovering all the relevant frequency spectrum information.[4]

The use of a relatively high-power picosecond mode-locked laser for resonant DFWM could simultaneously avoid thermal-grating interferences and increase data rates over pulsed lasers. Mode-locked Ti:sapphire is broadly tunable and can produce as much as 2 W of radiation with 2-ps pulses at ~82 MHz.[5] Picosecond four-wave mixing on sodium was previously reported,[6] but that study focused on grating properties in an optically thick flame. In this Letter we present an initial demonstration that short pulses from a mode-locked Ti:sapphire laser can induce normal DFWM signals in optically thin flames. In addition, although resonant DFWM is still in the development stage as a combustion diagnostic, we demonstrate that it is possible to use a cw mode-locked laser in this way to acquire previously unavailable turbulence spectra.

Our experimental layout is depicted in Fig. 1. The burner was a Perkin-Elmer aspirating unit fitted with a Meeker-type burner head. Various solutions of potassium chloride in water were aspirated into the air flow to provide controlled reproducible levels of atomic potassium in the flame. We used potassium in this exploratory research because it has strong lines near the peak of Ti:sapphire.

We used a Spectra-Physics regeneratively mode-locked Ti:sapphire laser equipped with a 2-ps Gires–Tournois interferometer.[5] We purged the cavity with the highest dry-nitrogen flow possible because there are many atmospheric absorption lines within the bandwidth of Ti:sapphire, including but not limited to the better-known water lines. Even small intracavity absorption can cause tuning discontinuities in the laser. We originally tuned to the $4^2S_{1/2}-4^2P^\circ_{3/2}$ transition in atomic potassium at 766.5 nm, but oxygen lines with 1% absorption closely surround 766.5 nm, and we experienced tuning discontinuities. We then chose the $4^2S_{1/2}-4^2P^\circ_{1/2}$ transition at 769.9 nm. This region has fewer atmospheric interferences, thereby giving some improvement in tuning. This laser produced ~900 mW of output when pumped with 7 W of energy from an argon-ion laser, with autocorrelation pulse widths near 1.4 ps (assuming a sech^2 pulse shape). The transform-limited bandwidth was ~0.5 nm, consistent with our monochromator (0.5-nm bandwidth) measurements.

With respect to Fig. 1(a), ~450 mW of power arrived at the DFWM setup. The two beam splitters (bs1, bs2) transmit 70% and reflect 30% of the laser intensity. Adjustable delay lines were inserted into all three beams by retroreflection from roof prisms

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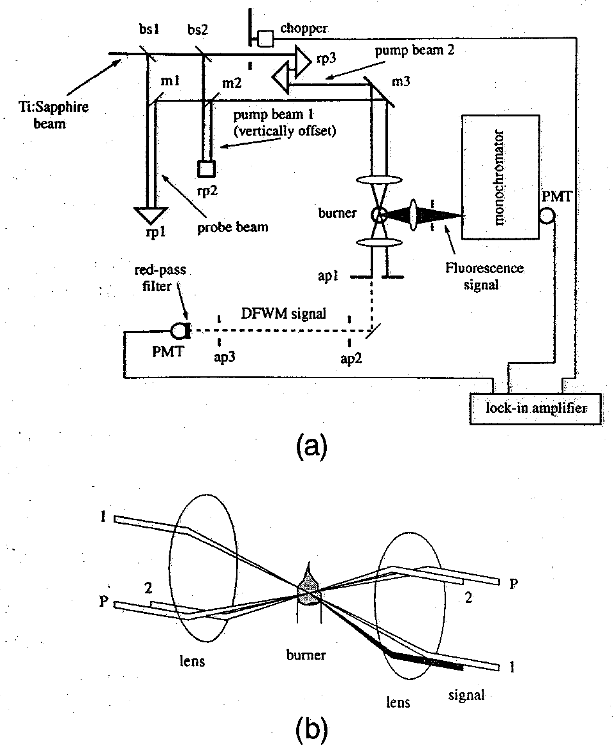

Fig. 1. Schematics of the DFWM setup.

(rp1, rp2, rp3) mounted on translation stages. Roof prism rp2 offset the beam downward, and mirror m2 was stationed below the probe beam. Pump beam 2 was mechanically chopped at ~1 kHz. We collimated and aligned all three beams in the far field, using matched drilled masks 0.63 cm × 0.63 cm, and then directed them through a lens to the flame position. The lens has a 7.5-cm focal length and gives a measured $1/e^2$ focal diameter of 116 μm. Final pulse temporal alignment was verified by simultaneous static autocorrelation in KDP at the lens focus. Focusing in the forward geometry [Fig. 1(b)] offers several advantages over the normal phase-conjugate arrangement: the three beams are immediately overlapped in the same location; it makes the autocorrelation scheme simple to perform; the intensities at the spot are then near the saturation intensity[7]; and this arrangement minimizes beam-steering effects. We wavelength tuned the laser by scattering some of the beam into the monochromator; then the flame was ignited, and we observed potassium fluorescence while tuning and simultaneously mode locking. Finally, when DFWM signals had been established, the laser could be tuned to maximize that signal.

Power levels in the three beams at the focal spot were $P(1) = 40$ mW, $P(2) = 39$ mW, and $P(P) = 60$ mW. To estimate roughly the saturation intensity for this non-steady-state regime, we set T_1 equal to the laser pulse width in the expression for I_{sat} given by Abrams *et al.*[7] This calculation gave an estimated ratio at line center of $I/I_{\text{sat}} \approx$ 3–4. This is not an accurate representation of saturation in this experiment, however, because the laser bandwidth is broad (100 times the potassium linewidth), and saturation in the wings of the line is more difficult to achieve.[8]

The DFWM phase-matching condition dictates that the signal scattered by pump beam 2 off a grating formed by pump beam 1 and the probe beam should exit at the fourth corner of the box [Fig. 1(b)]. We used confocal arrangement with a second matched lens. The array of beams could thus be recollimated through the drilled masks and the signal beam can be roughly located in space. An RCA 1P28 photomultiplier tube (PMT) was used to detect the signal, and we filtered spurious scatter, using an array of apertures (ap1, ap2, ap3). The PMT output was directed to a lock-in amplifier.

This DFWM arrangement gave maximum signal levels of ~140 mV with background scatter levels of 1–2 mV. In comparison, our fluorescence signal levels were ~100 μV with a 30-μV background.

Initial saturation results were acquired with a relatively large potassium chloride concentration (1.39×10^{12} cm^{-3}). The DFWM signal does not exhibit saturation, and this is likely due to the laser bandwidth. A best fit of the DFWM signal to the pump power gives a dependence of $I_{\text{signal}} \propto I_{\text{pump}}^3$ (correlation coefficient 0.997), consistent with expectations.[9]

Sensitivity curves are given in Fig. 2. To obtain these data we used solutions of varying potassium chloride concentration and observed the DFWM signal. Each data point in the figure is the average of from three to five DFWM measurements. An absorption measurement taken with a tungsten lamp was then performed to calibrate the data. The same potassium chloride solutions were used, and the flow rates of fuel, air, and solution were carefully matched between the DFWM experiment and the calibration. A curve fit to the data produces a quadratic signal dependence on concentration (correlation coefficient 0.990), consistent with expectations.[9]

The detection limit for this experiment, based on statistical variations in the data, is approximately 5×10^{10} cm^{-3}. This is not a particularly low number. We are aware of several problems that prevented lower detection limits. First, in our case the variations in data were not random; they were caused by laser wavelength drift with time. We typically saw the DFWM signal drop by 40% over the course of 30 min. A quick experiment demonstrated that his drift is most likely due to the fact that our pump laser did not have a beam-position stabilization system.

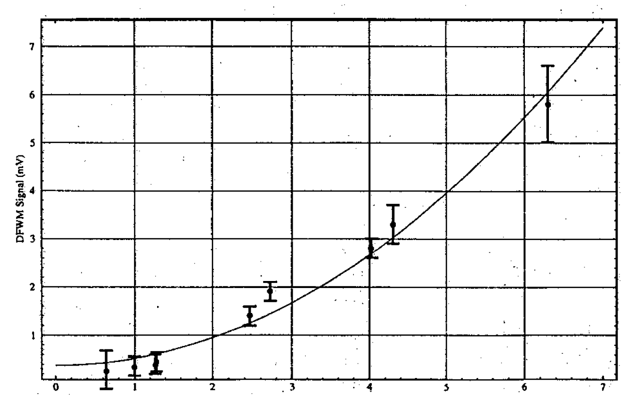

Fig. 2. DFWM signal versus K-number density (N). The signal scales with N^2.

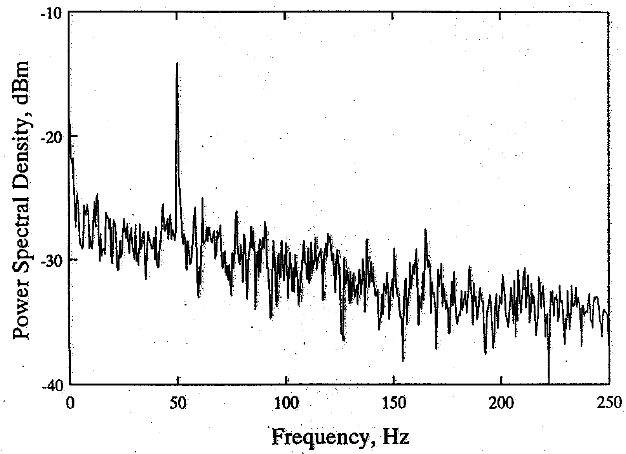

Fig. 3. Spectrum for an acoustic forcing frequency of 50 Hz.

If this drift were reduced, the detection limit could be reduced similarly. A second limitation in these preliminary results is that the low-concentration signals were artificially weak because we used the same high potassium concentration (1.39×10^{12} cm^{-3}, near optically thick) when tuning the laser wavelength. It is not appropriate to use such a high concentration when wavelength tuning on the DFWM signal because correct tuning will result in beam attenuation at the center of the line, giving a decreased DFWM signal.[10,11] Wavelength tuning is this high-concentration regime will produce larger DFWM signals when tuning is to the side of the line, but this will decrease the signal level at low concentrations. The obvious solution is to perform the final wavelength tuning on DFWM at low concentrations. Finally, our signal detection limit was set by spurious laser scatter and poor signal amplification. Scatter can be significantly reduced by the use of a spatial filter. We conclude that picosecond DFWM in flames behaves as expected, and there are several clear steps by which one can improve the measurement.

To demonstrate that we can detect the spectrum of a modulated flame, we mounted a small audio loudspeaker on the fuel–air line, and we drove it with a 50-Hz square wave. The loudspeaker induced a known disturbance in an otherwise laminar flame. The synchronously demodulated output of the lock-in amplifier was directed to a PC-based analog-to-digital system with a fast-Fourier-transform board. The power spectral density generated (see Fig. 3) clearly shows the 50-Hz modulation. The noise levels shown are due primarily to the fact that we did not use an instrumentation amplifier, and the low signal levels contributed to analog-to-digital noise. Our modulation rates (mechanical chopper and acoustic loudspeaker) were all fairly slow because we used borrowed equipment, none of which was fast. The choice of 50 Hz in no way implies a bandwidth limitation to the basic idea. This experiment clearly demonstrates the utility of a cw-based system for detection of turbulence statistics. Such a system can also be used for investigations of flame stability, acoustics and rapid transients, such as ignition and extinction.

The results presented here demonstrate a concept, but it should be possible to do much better. An effective way to optimize I/I_{sat} for DFWM would be to broaden the pulse to 50 ps, thus narrowing the bandwidth. Only ~1% of the laser energy overlaps an atmospheric pressure potassium line when 1.4-ps pulses are used. Longer pulses should produce larger signals since the effective intensity will be increased, and this is a currently unsaturated nonlinear process. Wavelength drift will need to be reduced with a pump-beam-positioning servo control. With respect to procedures, the laser will be wavelength tuned on the DFWM signal, in low-concentration regimes. In addition, we are currently setting up a spatial filter to reduce the background scatter levels. We can then route the PMT output directly to an instrumentation amplifier and then to the analog-to-digital system.

In conclusion, picosecond DFWM offers the possibility of a background-free determination of species and temperature that is less dependent on the collisional environment. Moreover we have demonstrated that mode-locked laser-based diagnostics can be used to acquire important turbulence spectra that have been unavailable until now.

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