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Degenerate Four-Wave Mixing in a Methane/Air Flame Using a Regeneratively Mode-Locked Ti:Sapphire Laser

M.A. Linne
Division of Engineering, Colorado School of Mines
Golden, CO 80401
(303) 273-3609

G. J. Fiechtner
Center for Combustion Research, University of Colorado
Boulder, CO 80309-0427

The potential of resonant Degenerate Four-Wave Mixing (DFWM) for measurements of major and minor species concentrations and temperature in reacting flows has been established by several groups[1]. Spatial resolution has been demonstrated, as has two-dimensional planar imaging. Perhaps most significantly, large signals have been observed at moderately high pressures (1100 Torr)[2], establishing the potential for measurements inside practical combustion systems. As a result, a good deal of research is currently devoted to understanding the fundamental physical aspects of the technique.

Degenerate Four-Wave Mixing is a third order nonlinear optical process. Three beams at the same wavelength (and typically from the same laser) are overlapped in space, according to phase matching constraints, and a fourth signal beam is produced. The nonlinear polarization of the medium is expressed in terms of a third order susceptibility [$X^{(3)}$] together with the vector interactions of the 3 input waves [3] $\mathbf{E}_1$, $\mathbf{E}_2$, & $\mathbf{E}_p$:

$$\mathbf{P}_{NL} = 1/2 \{ A (\mathbf{E}_1 \bullet \mathbf{E}_p^*) \mathbf{E}_2 + B (\mathbf{E}_2 \bullet \mathbf{E}_p^*) \mathbf{E}_1 + C (\mathbf{E}_1 \bullet \mathbf{E}_2) \mathbf{E}_p^* \} + \text{c.c.} \qquad (1).$$

The three input beams are labeled "pump beam 1", "pump beam 2", and the "probe beam". The coefficients A, B and C are related to tensor components of the susceptibility $X^{(3)}$. $\mathbf{E}_p^*$ is the complex conjugate of $\mathbf{E}_p$. The nonlinear polarization represented in equation (1) is then used as a source term in the wave equation. Phase matching between the four wave vectors in the form : $\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_p + \mathbf{k}_c$ {$\mathbf{k}_c$ = the signal (conjugate) wave vector} is required in order to couple the nonlinear polarization into the electric fields. A portion of the first term in equation (1), $A (\mathbf{E}_1 \bullet \mathbf{E}_p^*)$, describes a spatial interference pattern which is imprinted upon the medium via the susceptibility-related term "A". It could be due, for example, to the Kerr effect, or to absorption (resonant DFWM). The imprinted interference pattern is thus a standing, sinusoidal material grating. The remaining portion of the first term in equation (1) , $\mathbf{E}_2$, then represents scattering of pump beam 2 from the grating. The second term in equation (1) is similar to the first, and the third term does not contribute to resonant, single-photon DFWM.

There is a strong resonant enhancement of the DFWM signal when the laser is tuned to an atomic or molecular absorption. For the case of broadband excitation (as we have used in this work), the laser creates both a population (amplitude) grating at line center, and a dispersive (phase) grating in the wings of the line. Farrow et al[4] have extended the Abrams and Lind[5] perturbative, steady-state, two-level model to the case for low intensities (relative to the saturation intensity) and for broadband excitation (integrated over the absorption profile). Their result predicts that resonant DFWM signals, for the assumptions involved, will be proportional to number density squared. If we assume that the three input beams are of equal intensity, then the signal should scale with intensity cubed.

Recent studies in cells at various pressures and with various collision partners, using pulsed lasers (nanosecond widths), have shown that line broadening arguments can not always account for the observed collisional effects on DFWM signals[6,7]. It is generally agreed that $X^{(3)}$ wave mixing does occur, but energy can be transferred by collisional processes from population fringes into both temperature and pressure waves, giving rise to a strong thermal (phase) grating[8]. Signal interference from a thermal grating can make it very difficult to relate signal levels to number density. Detailed measurements, therefore, require the elimination of thermal grating interferences when they do occur. It has been known for some time that thermal gratings can be avoided by pumping with crossed polarization[9,10]. Rakestraw[10] has also pointed out that thermal gratings can be avoided by using picosecond pulses. Gasdynamic effects in flames of interest occur over several hundred picoseconds, so a measurement at about 50 ps or less would not be subject to thermal grating interferences. This is one of several reasons for doing DFWM the picosecond regime.

Our experimental layout is depicted in Fig. 1. The burner was a Perkin-Elmer aspirating unit fitted with a Meeker type burner head, operated near stoichiometric fuel/air ratios in this case. Various solutions of KCl in water were aspirated into the flow in order to provide controlled, reproducible levels of atomic potassium in the flame.

We used a Spectra-Physics regeneratively mode locked Ti:Sapphire laser, equipped with a 2 ps Gires-Tournois interferometer[11]. This laser produced about 900 mW of output when pumped with 7 W from an Ar:Ion laser, with autocorrelation pulse-widths around 1.4 ps (assuming a $sech^2$ pulse shape). The transform limited bandwidth is about 0.5 nm. The laser was tuned to the $4^2S_{1/2}$ - $4^2P^o{}_{1/2}$ transition in atomic potassium at 769.9 nm.

Referring to figure 1a, about 450 mW arrived at the DFWM setup. The two beam splitters transmit 70% and reflect 30% of the laser intensity. Adjustable delay lines were inserted into all three beams by retro-reflection from roof prisms mounted on translation stages. Roof prism rp2 offset the beam downward, and mirror m2 was stationed below the probe beam. Pump beam number 2 was considered the reference beam for alignment. It was mechanically chopped at about 1 kHz and it was fitted with a set of roof prisms capable of up to 530 ps delay. We collimated and aligned all three beams in the far-field using drilled masks, and then directed them through a lens to the flame position. Temporal alignment was verified by simultaneous, static autocorrelation in KDP at the lens focus. The lens has a 7.5 cm focal length and gives a measured $1/e^2$ focal diameter of 116 µm. Focusing offers several advantages : the three beams are immediately overlapped in the same location, giving excellent spatial resolution in the transverse direction, it makes the pulse autocorrelation scheme simple to perform, and the intensities at the spot are then near the saturation intensity[5].

Power levels in the three beams at the focal spot were P(1) = 40 mW, P(2) = 39 mW, and P(P) = 60 mW. This gave an estimated ratio at line center around $I / I_{sat} \approx 3 - 4$. This is not an accurate representation of saturation in this experiment, however, because the laser bandwidth spans the entire line. Saturation in the wings of the line is more difficult to achieve[12].

The all-forward geometry we used is similar to BOXCARS (Fig. 1b). The DFWM phase matching condition dictates that the signal scattered by pump beam 2 off of a grating formed by pump beam 1 and the probe beam should exit at the fourth corner of the box. We used a confocal arrangement with a second, matched lens. The array of beams could thus be be re-collimated through the drilled masks and the signal beam can be roughly located in space. We filtered spurious scatter using an array of apertures. The signal was detected with a photomultiplier tube (PMT), and the PMT output was directed to a lock-in amplifier. We simultaneously monitored fluorescence at 90 degrees to the beam overlap region.

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

Initial saturation results indicate that a best-fit to the data gives a power law dependence of $I_{signal} \alpha I^3$, consistent with expectations. We did not reach full saturation, likely because of laser bandwidth. Solutions of varying KCl concentration were used to acquire sensitivity curves. An absorption measurement using a tungsten filament lamp was then performed to calibrate the data. The same KCl 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 does not produce a quadratic signal dependence on concentration, as one would expect. This is most likely due to beam absorption at higher concentrations[13,14]. If the high number density data are ignored, the remaining low concentration data fit a quadratic well. An important conclusion is that the measurement offers good sensitivity, limited primarily by the interference of laser scatter.

Finally, we mounted a small audio loud speaker on the fuel / air line, and we drove it at 50 Hz with a frequency synthesizer. This induced a known disturbance in an otherwise laminar flame. The output of the lock-in amplifier was directed to a PC based A/D system with an FFT board. The FFT's generated (see Fig. 2) clearly show the 50 Hz modulation. This simple experiment demonstrates the utility of an effectively cw based system for detection of turbulence statistics. The noise level represents A/D noise caused by low signal levels. A high quality instrumentation amplifier would improve the signal/noise ratio considerably.

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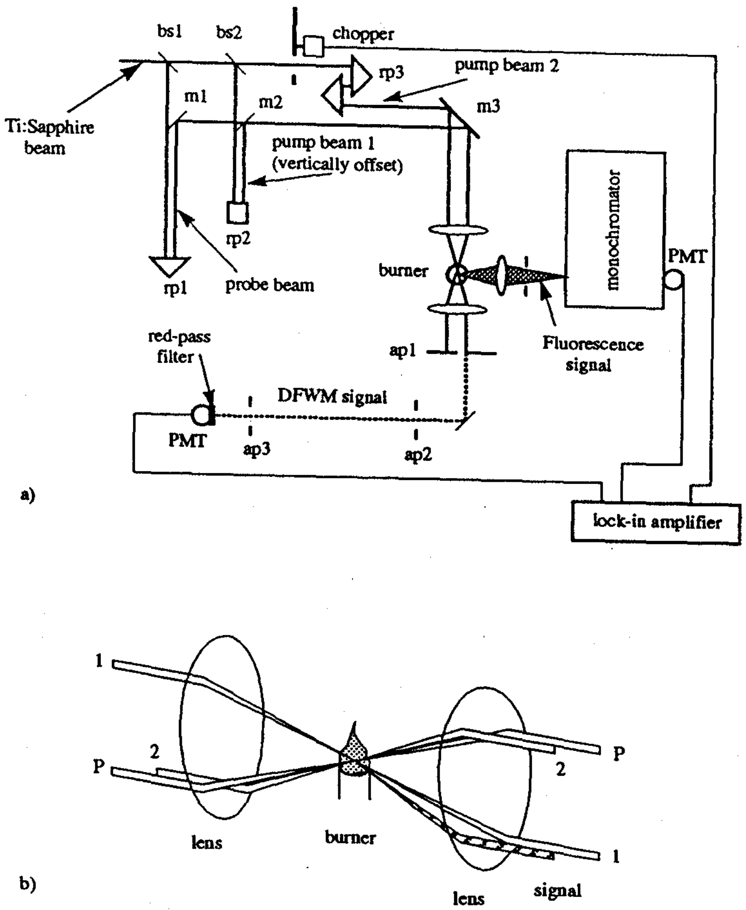

Figure 1. DFWM layout.

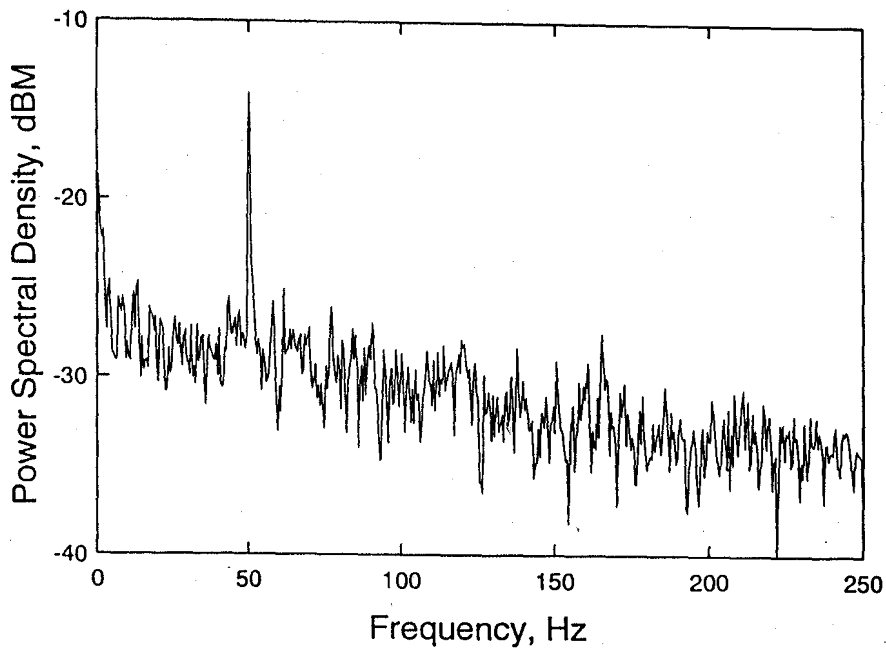

Figure 2. Frequency spectrum of 50 Hz modulated flame.