

# Two-Dimensional Pump/Probe Imaging in Reacting Flows

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## *Abstract*

We have demonstrated that picosecond Pump/Probe spectroscopy could be used to acquire quantitative, two-dimensional images of species in flames, using a loss-modulation based, two-dimensional phase-sensitive detection system mounted in front of a CCD camera. This system is currently limited to detection of 1% modulation depth, but it can be significantly improved upon. The instrument allows high data collection rates, and it provides a quenching independent signal without the need for calibration. The signal is imbedded in a coherent laser beam, making it possible to perform optical processing before the camera.

Here we describe Pump/Probe spectroscopy, describe our imaging technique, present current imaging results and then discuss what needs to be done to improve the technique.

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## Introduction

Two-dimensional imaging of radical species concentrations in reacting flows (e.g. OH, CH, NO etc.) has yielded a large amount of useful information for modelers. As one example, Planar Laser Induced Fluorescence (PLIF) has become the technique of choice for turbulent flame studies (Hanson, 1986). While PLIF has proven invaluable, there are a number of data manipulations required in order to make the measurement quantitative, such as the collisional quenching correction. In non-premixed, turbulent flames it is difficult to quantify the collisional environment within each pixel area in the flow. This has proven to be a significant limitation to the PLIF technique, but there are other required corrections. Since LIF is proportional to intensity, and laser beams have spatial dependence, it is necessary to measure the beam profile and to account for it. Each camera has responsivity which varies from pixel to pixel as well, and so it is necessary to account for this instrument response. There have, in fact, been very few quantitative PLIF results reported, even in steady laminar flames (Smyth, 1994).

We have recently demonstrated several new diagnostic techniques based upon picosecond, mode-locked Ti:sapphire lasers (M. A. Linne and G. J. Fiechtner, 1994; G. J. Fiechtner and M. A. Linne, 1994). The principal advantages of these techniques include : 1) the fact that picosecond pulse diagnostics are not expected to be strongly affected by the collisional environment, and 2) the high repetition rates of mode-locked lasers (70 - 100 MHz) can make it possible to observe all the time scales of importance in turbulence, or to observe rapid (sub-millisecond) transients (e.g. ignition). The Pump/Probe technique offers the potential for determination of absolute number density, free of corrections and without the need for calibration (G. J. Fiechtner *et al.*, 1995). These techniques, therefore, have the potential to overcome several of the difficulties associated with PLIF.

## Pump/Probe Spectroscopy

In Pump/Probe spectroscopy (see Fig. 1), the pump beam is modulated, and it is crossed with the probe in an absorbing sample. The pump modulation frequency is impressed on the resonant molecules. These molecules then modulate the probe via absorption and stimulated emission. The probe modulation is measured with a photodetector and lock-in amplifier (phase sensitive detection), and this measurement can be directly related to the number density of absorbers in the overlap between the two beams according to the following expression (G. J. Fiechtner *et al.*, (1995) :

$$\alpha_{\text{MOD}} = \frac{g_2}{g_1} \left( 1 + \frac{g_2}{g_1} \right) \left[ \ln \left( \frac{1}{\sqrt{2}-1} \right) \right]^2 \frac{c^4 A_{21}^2 P_{\text{AVE}}^{\text{PUMP}} N_T L}{16\pi D^2 h \nu_{12}^5 f^L (\Delta \nu_{1/2}^L)^2} \quad (1).$$

where:

$g_1$	= ground state degeneracy,	$P^{\text{PUMP}}$	= average laser power in pump beam,
$g_2$	= excited state degeneracy,	$A_{21}$	= Einstein coefficient for spontaneous emission.
$D$	= interaction focal diameter,		
$L$	= interaction length,		
$f^L$	= laser repetition rate,		
$\nu_{12}$	= 1 -> 2 transition frequency,		
$\Delta \nu_{1/2}^L$	= laser bandwidth (FWHM),		

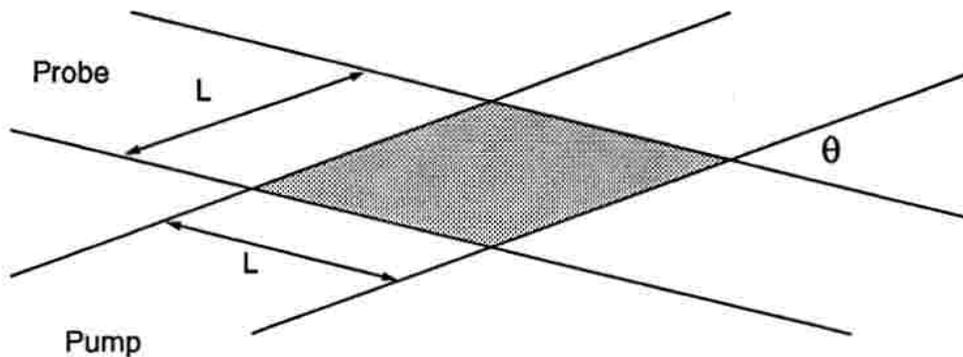


Figure 1. Schematic of Pump/Probe experiment.

The development of equation (1) assumes optically thin conditions, operation in the linear absorption regime, a 3-level absorber (potassium in our case), transform-limited pulses, a temporal top-hat pulse profile, and broad bandwidth with respect to the absorption linewidth.

Measurement of  $\alpha_{MOD}$  is straightforward. The probe beam is directed into a photodiode and the photodiode signal is connected to a lock-in amplifier. Next : 1) The chopper is removed from the pump and placed in the probe, and the lock-in amplifier signal is recorded, giving 100% modulation depth. 2) The chopper is removed altogether and the lock-in amplifier signal is recorded, giving 0% modulation depth. 3) The chopper is placed in the pump beam, and the lock-in amplifier signal is recorded. That gives a relative  $\alpha_{MOD}$  (relative to 100% & 0%) for use in equation (1). We have previously seeded potassium into a methane/air flame to demonstrate that Pump/Probe delivers an absolute potassium number density at very rapid detection rates (G. J. Fiechtner and M. A. Linne, (1994)).

The stated assumptions are not appropriate for more complex absorbers, such as the free radicals of interest to combustion researchers. Hence, we do not expect to find an equation for free radical number density based upon modulation depth as simple as expression (1), although we do expect to find a relationship relating an experimentally measured modulation depth to the number density of absorbers. Development of such a relationship is a current topic of research.

### ***Two-Dimensional Phase Sensitive Detection***

Here we demonstrate that this same technique can be used to acquire two-dimensional images of number density in flames. To perform this experiment, we have expanded the pump beam into a sheet using cylindrical optics (see Fig. 2). The probe is then up-collimated into a large diameter, gaussian cross-section beam which overlaps the sheet in the flame. It is then necessary to develop a two-dimensional analog to the lock-in amplifier used in the single-point experiments. Two-dimensional phase sensitive detection could be accomplished in software (similar to gated integration), but that requires post processing of saved images. In order to achieve good limits of detection, a very large number of images would have to be averaged. That would limit this technique to steady flows. Instead, we have chosen to demonstrate that it is possible to demodulate the probe image in front of a CCD camera.

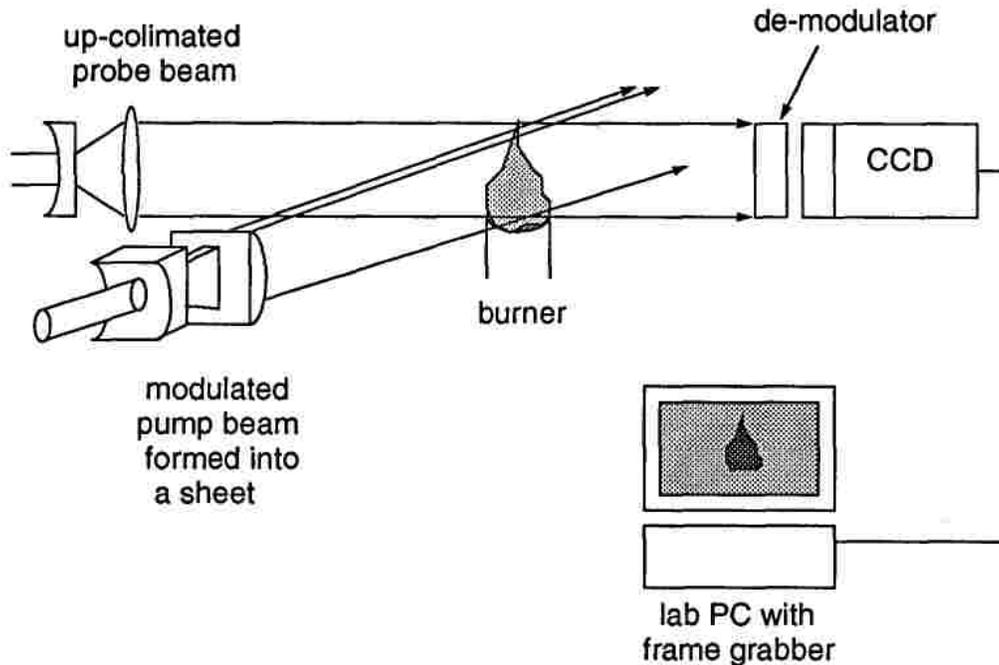


Figure 2. Schematic of two-dimensional Pump/Probe implementation.

In phase sensitive detection, some input to the experiment (the pump beam here) is modulated (say at  $\omega_m$ ). That system input produces an output signal (here - a modulation on the probe beam) induced by the variable of interest (absorber atoms). The modulated signal has the same frequency as the input, but it can have a phase shift and is typically imbedded in a relatively large background consisting of noise components covering a broad range of frequencies ( $\omega_i$ , see Fig. 3). In a lock-in amplifier, the total signal (modulated signal of interest + large background) is first demodulated by a mixer. This occurs by multiplication of the total signal with a sinusoid at the chopper frequency :

$$e_i \times e_m = 0.5 \sum_i (E_i \times E_m) [ \cos(\omega_i - \omega_m)t - \cos(\omega_i + \omega_m)t ] \quad (2),$$

producing sum and difference frequency terms for all portions of the total signal. The difference term is at zero frequency only for the modulated signal of interest (since  $\omega_i$  is the same as  $\omega_m$  in that case). That small portion of the total signal is "mixed down" to dc because it has the same frequency as the chopper. The output of the mixer is then routed through a low-pass filter which eliminates the sum frequency term of the modulated signal of interest, and any other components (e.g. the large background) which fall outside the bandwidth (user selected) of the filter. The remainder of the signal carries information about the variable of interest (contained in the sideband which passes through the low-pass filter).

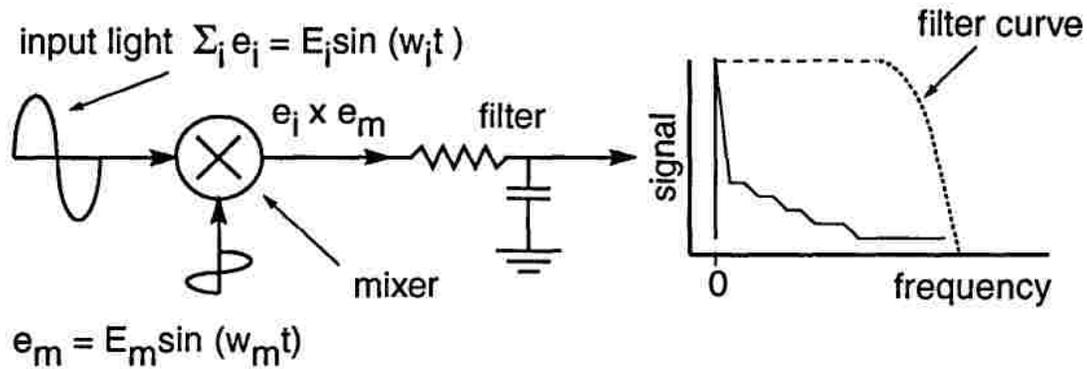


Figure 3. Schematic of a standard phase-sensitive detection system.

Mixers themselves can be thought of as amplifiers in which the amplifier gain is modulated at the chopper reference frequency. The amplifier output is the linear product of the input and the gain (sinusoidal), so this performs the multiplication just described. For imaging, it is possible to accomplish the same task using optical loss (by polarization modulation) or gain (by use of a micro-channel plate or photocathode) directly in front of the camera pixels. If, for example, the reference frequency is used to modulate loss, the optical signal arriving at each pixel is the linear product of the input and the reference waveform. Here we have used liquid crystals to perform this task, primarily because they are inexpensive and our goal was simply to demonstrate the concept. It remains to filter this demodulated total optical signal, accomplished here by the integrating behavior of the pixel itself together with the camera framing rate of 30 Hz. This is not necessarily the optimum filter behavior, but it proved sufficient for the present demonstration.

One important difference between our two-dimensional, optical phase sensitive detection system and a typical lock-in amplifier is that the demodulator is capable of reducing the light level to zero and then driving it up to some maximum level characteristic of the system (i.e. the optical signal will not go negative). That imposes a dc offset at the electrical input (see Figure 4), and hence the product of the reference and signal contains a non-negligible dc level which is

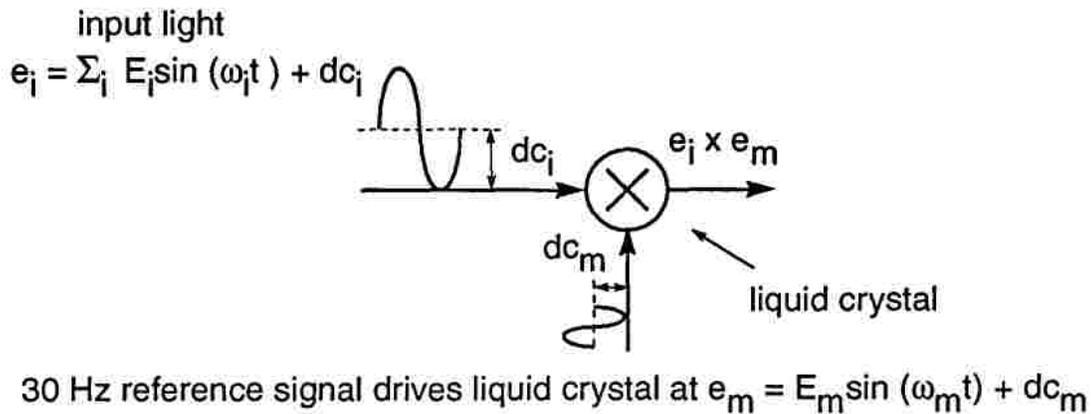


Figure 4. Schematic of optical mixing process.

summed with the demodulated signal. It is therefore necessary to subtract this dc component from the total.

Our liquid crystals can modulate light up to about 200 Hz. For this reason, we chose to chop the pump beam and to drive the liquid crystal demodulator at 30 Hz. The sinusoidal drive for the crystal was synthesized using the chopper output. Because of the slow modulation frequency, there was effectively no phase difference between the reference and signal. The demodulation then occurs via multiplication of the input with the reference signals resulting in the following expression :

$$e_i \times e_m = \sum_i [\{dc_i \times dc_m\} + \{dc_i \times E_m \sin(\omega_m t)\} + \{dc_m \times E_i \sin(\omega_i t)\} + \{E_i \times E_m [0.5 \cos(\omega_i - \omega_m)t - 0.5 \cos(\omega_i + \omega_m)t]\}] \quad (3).$$

[where subscript i represents the optical signal and the subscript m represents the liquid crystal]

Here  $\omega_i = \omega_m = 2\pi \times 30 \text{ (s}^{-1}\text{)}$  for the signal of interest. For that signal, the difference term  $\{\cos(\omega_i - \omega_m)t\}$  goes to one, and the sum term  $\{\cos(\omega_i + \omega_m)t\}$  is a second harmonic of the 30 Hz. The camera framing rate is the same as the chopper rate, 30 Hz, but the signal demodulation and the chopper reference are synchronous. They are not synchronous with the camera. As such, the camera acts simply as a low-pass filter which does not interact with the mixing process. Such a camera sampling filter curve is shown in Figure 5 (for one camera frame). It has zeros at the framing rate harmonics, which effectively filters all the  $\omega$  and  $2\omega$  terms in equation (3). What's left is  $\{dc_i \times dc_m\}$ , the dc offset problem, and  $\{0.5 E_i \times E_m\}$ , our signal. A typical lock-in amplifier operated at 30 Hz would require a long time constant to provide good signal-to-noise ratios (SNR). That is also the case here, where we integrate over or 8 - 9 seconds (256 images). To accomplish this, we store a running average on the frame grabber, and average each new frame with that image as it is acquired. This is in effect exactly what occurs in the filter of a lock-in amplifier.

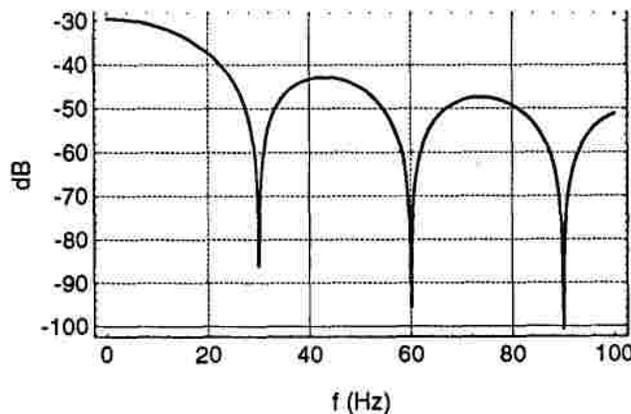


Figure 5. Camera framing filter curve.

Finally, it remains to subtract the dc offset from the image. In order to do that we modulate the crystal at 300 Hz, beyond its ability to modulate light but preserving the neutral density value. That eliminates the  $\{E_i \sin(\omega_i t) \times E_m \sin(\omega_m t)\}$  term but preserves the  $\{dc_i \times dc_m\}$  term. We store that dc image and subtract it from the demodulated image, giving us then the remainder, the signal of interest :  $\{E_i \sin(\omega_i t) \times E_m \sin(\omega_m t)\}$ .

### ***Experimental Results***

For the experiments reported here, we used a Perkin-Elmer aspirating burner fitted with a Meeker-type burner head (see G. J. Fiechtner and M. A. Linne, (1994) for experimental details). A solution of potassium chloride in de-ionized water was aspirated into the burner. We used a Spectra-Physics Tsunami Ti:sapphire laser operated at 60 ps and tuned to the  $4^2S_{1/2} - 4^2P^o_{3/2}$  of atomic potassium at 766.49 nm (near the peak of the Ti:sapphire fundamental wavelength). The 130 mW pump was spread into a sheet approximately 2.5 cm tall (the flame height) and 1 mm thick. The pump and probe irradiances were smaller than their saturation values. The low-intensity probe was spread into a cylinder about 2.5 cm in diameter, and it was overlapped with the pump at 90°. That beam was displayed on a screen, and it was then viewed through the liquid crystal by a Javelin CCD camera. The image was acquired and manipulated with a Sharp GPB-1 frame grabber mounted in a personal computer.

Figure 6 a. contains the image of the probe which was visible on the screen while the flame was on and absorbing. Note that the center of the gaussian probe was off of the flame centerline, as shown by the shadow of the burner at the bottom of the photograph. Figure 6 b. contains the demodulated image after the dc term has been subtracted. We were unable to assess the full modulation depth in these experiments, due to the auto-gaining nature of this borrowed camera (the auto gain loop was too slow to affect demodulation). It was therefore not possible to obtain an absolute number density using equation (1). We know, however, that the average concentration of potassium represented by the image is just under  $10^{12} \text{ cm}^{-3}$ , which is nearly optically thin. One can, in fact, see that there is no probe absorption visible in Figure 6 a. It is not possible to detect lower concentrations than this with the current system because an 8-bit digitizer is incapable of resolving better than about 1% modulation in a large background. The edges visible in Figure 6b are the edges of the mirror used to direct the image onto the screen, and they contain some scattered modulated pump light (which can be eliminated by spatial filtering). Finally, we independently de-tuned the laser and then aspirated pure water through the burner in order to observe no image, thereby assuring that we were indeed imaging potassium.

### ***Future Work***

The entire system just described was devised to accommodate the slow optical modulation speed of the liquid crystals. If we were using electro-optics, for example, we could demodulate the probe images at frequencies in the MHz regime, which would allow the job to be accomplished within one camera frame. The background problem remains, and it imposes a high detection limit because an 8 bit system can only resolve a modulation depth of 1%. In order to overcome this problem in Pump/Probe spectroscopy, it would be necessary to acquire the dc images simultaneously (in a second detector array) and subtract the two images as analog signals before digitizing. Alternatively, one could use Polarization Spectroscopy (see for example K. Nyholm *et al.*, 1993), Antiresonant-Ring Transient Spectroscopy (see for example R. Trebino



a.

b.

Figure 6. a. Probe beam background plus modulated image (not visible).; b. Demodulated image extracted from 2a.

and C. Hayden, 1991) or Degenerate Four-Wave Mixing (D. J. Rakestraw *et al.*, 1990, M. A. Linne and G. J. Fiechtner, 1994). These are all crossed-beam, background-free techniques. The low signals produced would require the use of a photocathode/image intensifier system as the demodulator instead of a polarization technique, and this would introduce some reduction in spatial resolution as a trade-off.

We have made performance estimates for various camera geometries including liquid crystal demodulation, electro-optic demodulation, and demodulation using the photocathode preceding the microchannel plate in an image intensifier. For combustion imaging to be viable, it will be necessary to detect a modulation depth of  $10^{-8}$ . The dc background level encountered in basic Pump/Probe will not allow this detection limit. The average electron pool per pixel in a CCD is 300,000, giving a shot noise value of 547. If the dc value of the signal exceeds the magnitude of the modulated component by more than a factor of 547, the data can not be recovered reliably. Even if this were not the case, an 8 bit board can not resolve better than 1% modulation depth. Higher resolution boards are available, but the dc component will have to be reduced by a factor of  $2 \times 10^5$  in order to overcome the electron-well problem. If one of the background-free techniques can remove  $10^4$  of the background, then the remaining dc component can be handily removed by the use of phase retardation for intermittent camera frames. If the demodulation phase is retarded by  $180^\circ$  in that intermittent frame, then the image stored will be *dc - demodulation*, vs *dc + demodulation* for the phase =  $0^\circ$  frames. Post processing will allow one to subtract the remaining dc term from these two.

Clearly, the next step is to set up a single-point pump/probe experiment and to test these background-free techniques. The results of that work will demonstrate what should be possible for imaging of radical species.

### ***Conclusions***

PLIF is a much more mature technique than the Pump/Probe imaging approach discussed here. All the same, it might be useful at this point to compare the attributes of both. PLIF provides an 8 ns snapshot of a free radical distributions in a reacting flow. The camera is, in effect, shuttered by the laser pulse, providing excellent time-resolution. Furthermore, LIF is a very sensitive, background free technique, so high quality images are acquired directly. These are very useful attributes. As we mentioned in the introduction, however, there are a number of data manipulations required in order to make the measurement quantitative, such as the collisional quenching correction. In non-premixed, turbulent flames it is difficult to quantify the collisional environment within each pixel area in the flow. This has proven to be a significant limitation to the PLIF technique, but there are other required corrections. Since LIF is proportional to intensity, and laser beams have spatial dependence, it is necessary to measure the beam profile and to account for it. Each camera has responsivity which varies from pixel to pixel as well, and so it is necessary to account for this instrument response. There have, in fact, been very few quantitative PLIF results reported, even in steady laminar flames (Smyth, 1994).

This new Pump/Probe technique is expected to be insensitive to the collisional environment, and it has the potential to deliver quantitative determinations of number density with no corrections or calibrations. Pixel response and beam profile are not an issue because each pixel behaves as an individual photodetector, determining a local modulation depth based upon 100% and 0% modulation for that location (beam and pixel), as described above. That point is driven home by the fact that in Figure 6 a., the probe beam was off-center. The wings of the beam contained the modulation, not the core of the beam. Despite this fact, the demodulated image is clean and uniform. Moreover, the modulated signal is contained in a laser beam, which can be subjected to optical processing before it is recorded by the camera. This allows spatial filtering, for example. Since the data rate is high, one can imagine acquiring images at the camera framing rate. This would, of course, necessitate data storage to tape and post-processing. The down-side to this idea is that the camera frames at 30 Hz, and at least in the short term we'd like to use as much of the pixel integration time as possible, for improved SNR. That means that the flow field under study has to have characteristic times on the order of  $< 0.3$  sec, or the images will be blurred. Images in unsteady, high speed flows would be of low quality. It is possible to purchase cameras which frame in the kHz regime, with image intensifiers. In the future it may be possible to do Pump/Probe or similar imaging using one of these.

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