DEVELOPMENT OF HIGH REPETITION RATE NO PLANAR
LASER INDUCED FLUORESCENCE IMAGING

DISSERTATION

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ABSTRACT

This thesis has documented the development of a MHz repetition rate pulse burst laser system. Second harmonic and third harmonic efficiencies are improved by adding a Phase Conjugate Mirror to the system. Some high energy fundamental, second harmonic, and third harmonic burst sequences consisting of 1 – 12 pulses separated in time by between 4 and 12 microseconds are now routinely obtained. The reported burst envelopes are quite uniform.

We have also demonstrated the ability to generate ultra-high frequency sequences of broadly wavelength tunable, high intensity laser pulses using a home built injection seeded Optical Parametric Oscillator (OPO), pumped by the second and third harmonic output of the pulse burst laser. Typical OPO output burst sequences consist of 6 – 10 pulses, separated in time by between 6 and 10 microseconds.

With third harmonic pumping of the OPO system, we studied four conditions, two-crystal Singly Resonant OPO (SRO) cavity, three-crystal OPO cavity, single pass two-crystal Doubly Resonant OPO (DRO) cavity and double pass two-crystal OPO cavity. The double pass two-crystal OPO cavity gives the best operation in burst mode. For single pass OPO, the average total OPO conversion efficiency is approximately 25%. For double pass OPO, the average total OPO conversion efficiency is approximately 35%.
As a preliminary work, we studied 532nm pumping of a single crystal OPO cavity. With single pulse pumping, the conversion efficiency can reach 30%.

For both 355nm and 532 nm pumping OPO, we have demonstrated injection seeding. The OPO output light linewidth is significantly narrowed. Some preliminary etalon traces are also reported.

By mixing the OPO signal output at 622 nm with residual third harmonic at 355 nm, we obtained 226 nm burst sequences with average pulse energy of ~0.2 mJ. Injection seeding of the OPO increases the energy achieved by a factor of ~2. 226 nm burst sequences with reasonably uniform burst envelopes are reported.

Using the system we have obtained, for the first time by any known optical method, Planar Laser Induced Fluorescence (PLIF) image sequences at ultrahigh (≥100kHz) frame rates, in particular NO PLIF image sequences, have been obtained in a Mach 2 jet.

We also studied the possibility of utilizing a 250 kHz pulsed Nd:YVO₄ laser as the master oscillator. 10-pulse-10-µs spacing burst sequences with reasonably uniform burst envelope have been obtained. The total energy of the burst sequence is ~2.5J.
Dedicated to my family
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CHAPTER 1

INTRODUCTION AND BACKGROUND

The application of optical diagnostic imaging techniques based on new laser technologies has developed enormously in recent years. In particular, the desire to study high Mach number – high Reynolds number flows has led to the development of several qualitative and quantitative flow imaging techniques, such as Planar Doppler Velocimetry (PDV), Particle Imaging Velocimetry (PIV), and Planar Laser Induced Fluorescence (PLIF). Such techniques require very short pulse duration sources, on the order of 10-100 nsec, in order to “freeze” instantaneous flow structure. While this is now rather common place, commercial pulsed solid state lasers have repetition rates in the range of 10-30 Hz, which is too slow, by many orders of magnitude to dynamically follow the evolution of high speed flow structure. To address this, a number of researchers have taken steps towards the goal of developing a coherent light source with high pulse energies and high repetition rates.
1.1 MHz Pulse Burst Laser

The pursuit of high repetition rate experimental diagnostics begins with the development of a suitable light source. A limited number of high repetition rate laser sources have been developed in the past few years.

Huntley [1994][1] and Grace et al. [1998][2] developed a repetitively Q-switched Ruby laser for holographic applications that could produce a burst of high energy pulses (order of 100s of mJ) with ~10 ns pulse widths over a timespan of ~140 microseconds at repetition rates up to 500 kHz. Operation-to-pulse of this laser system is possible at rates as high as 1 MHz. However, ruby lasers, which are 3 level systems, require exorbitantly high pump energies to achieve significant output. The resulting thermal load on the lasing medium necessitates intermittent operation of the laser from pulse burst to pulse burst (order of minutes), making them impractical for many applications.

Kaminski et al. [1999][3] and Hult et al. [2000][4] used a cluster of four individual double-pulsed Nd:YAG lasers. Each laser head was fitted with a double pulse option (DPO) where the “Q-Switch” can be pulsed twice in rapid succession during a single firing of the flashlamps, which is the optical pumping source for the Nd:YAG rod. This allows the extraction of up to two laser pulses from each cavity with a selectable time separation of ~100 us. With this system they can achieve bursts of 8 pulses with individual pulse energies of 270 mJ at 532 nm.

Reeves et al. [2000][5] used a Copper Vapor laser at ~10 kHz and ~5 mJ/pulse for PIV imaging. There are also many other high repetition rate laser systems, but the pulse energy is very low, less than 1 mJ per pulse, which is limited by the input power of the
system. For example, Crystal Laser Inc. has a commercial pulsed Nd:YAG laser with repetition rate of 250 kHz with ~100 uJ per pulse at 1064 nm. Simultaneous achievement of high repetition rate and high individual pulse energy has been a goal for high speed flow imaging researchers for many years.

In 1996, Lempert et al. [6] developed a Nd:YAG based pulse burst laser system in Princeton University. The “burst” concept is critical to high (MHz) rate imaging diagnostics, because the product of repetition rate and energy-per-pulse is constrained by the maximum average power thermal loading that solid-state lasing elements can tolerate. For example, the current maximum average power for commercially available Nd:YAG systems is approximately 15 Watts at 1064 nm (1.5 J at 10 Hz). This means that if a MHz repetition rate laser system were to be run continuously, material thermal considerations alone would limit the output energy to approximately 0.15 mJ per pulse. This is too low to be generally used for high speed flow imaging experiments. The key to the “burst” concept is, therefore, the reduction of the duty cycle, in order to achieve high energies in each individual pulse. Lempert et al. used a commercially available diode-pumped, CW Nd:YAG laser as the master oscillator. After amplification by a four-pass amplifier, the CW laser signal is sliced by a pair of electro-optic Pockel cells, similar to those used as Q-switches for standard solid-state laser systems, into a burst train. A few more single (or double) pass amplifiers were added into the system to make the pulse energy higher. With this system they could produce the burst sequences which contain between 1 and 99 pulses at up to 1 MHz with ~10 mJ/pulse at the fundamental output wavelength (1064 nm). Their system had a narrow spectral linewidth of ~60 MHz and was applied to Flow Visualization measurements in the boundary layer of a Mach 2.5 wind tunnel [6]. Lempert
et al. also applied this laser system for Rayleigh Scattering Measurements \cite{7}, and Wu et al. used the same system to visualize the shock wave/ boundary layer interaction over a 14-degree wedge in a Mach 2.5 flow \cite{8}.

Wu et al. developed a 2\textsuperscript{nd} generation pulse burst laser system with pulse energies on the order of 100 mJ at 1064 nm and 10s of mJ at the second harmonic wavelength of 532 nm. They applied it to Flow Visualization measurements of shock structures \cite{9} and shock evolution \cite{10} in a supersonic flow.

As part of this thesis, Lempert et al. \cite{11, 12} Thurow et al. \cite{13}, and Jiang et al. \cite{14} developed the parallel 2\textsuperscript{nd} generation pulse burst laser system which will be described in Chapter 2. The use of this system for PDV \cite{15} and PLIF \cite{14} measurements of high speed flow will be discussed in Chapter 2 and Chapter 4.

1.2 Experimental High-repetition rate Flow Imaging Techniques

A number of planar techniques have been developed over the last few decades, with the potential to be adapted to high-repetition rates. Shadowgraph and schlieren imaging are well known techniques that have been used for over a 100 years to provide qualitative visualizations of density changes within fluid flows. A number of researchers [e.g. Sarohia and Massier \cite{16} and Mahadeven et al. \cite{17, 18}] have adapted this classical technique to high-speeds using high framing rate film cameras. Although useful, these techniques are still limited by the spatially integrated, qualitative nature of schlieren systems.
More recently, researchers have employed laser sheet scattering flow visualization techniques, where a laser beam is formed into a sheet, directed through the flow field and light scatters from molecules/particles in the flow. Methods of particle seeding can vary greatly depending on the nature of the flow.

Particle imaging velocimetry (PIV) uses two time-correlated images of a particle-seeded flow and uses image processing to determine the velocity of the flow across the plane. This technique has been developed into a quite robust and reliable system for low-speed flows with many commercial systems available. The main challenge in any PIV system is proper seeding of the flow, which requires particles small enough to follow the flow, but large enough to scatter sufficient light to the cameras. A uniform distribution of particles throughout the flow is also required for optimum results. Application to high-speed flows has been constrained by the increased demands on particles seeding. Initial development of a MHz rate PIV system using a pulse burst laser of similar design has been conducted by Wernet and Opalski \[19\], with the assistance of the OSU group.

Planar Doppler velocimetry (PDV) \[20\] is a similar technique, in that it has the ability to measure the velocity across a plane in the flow field. PDV determines the flow velocity by measuring the Doppler shifted frequency of light as it is scattered by moving particles in the flow. The Doppler shift is measured using a molecular filter, whose absorption characteristics are a function of frequency. Unlike PIV, PDV does not require the visualization of individual particles; rather, it can determine the velocity of light scattered by groups of particles. This is particularly advantageous in high-speed flows where continuous particle seeding (i.e. product formation) can be achieved with much less difficulty. While the spatial resolution of PIV is limited by the size of the cross-
correlation window (typically 32 x 32 pixels), PDV can resolve smaller scales of velocity and approach resolutions on the order of a single pixel. In addition, PDV measurements can be made for each laser pulse, whereas PIV requires 2 time-separated images of the flow to calculate velocity. These characteristics make PDV a good technique for adaptation to MHz rates using the pulse burst laser. The development and application of MHz rate PDV is the subject of Chapter 2.

Another quantitative technique potentially suited for MHz rate adaptation is molecular Rayleigh scattering. Molecular Rayleigh scattering\[^7\] can be used to measure the density of the flow, but the signal is relatively weak and higher pulse energies are required. This problem can be partially circumvented by using gases with higher scattering cross sections, but this approach is often impractical. While it will be discussed in Chapter 5, Dr. Daniel Den Hartog, a senior research staff member in the plasma physics group, Department of Physics, at the University of Wisconsin Madison, has the goal of measuring Thomson Scattering by using the pulse burst laser system with ~1J individual pulse energy.

In this work, we have targeted NO based Planar Laser Induced Fluorescence (PLIF) as the main experimental technique. PLIF, which can either be qualitative or quantitative depending on the experiment, utilizes the fluorescence of individual molecules contained in the flow and is especially suited for measurements in reacting flows where the production of products and depletion of reactants can be measured. However, for MHz repetition rate NO PLIF imaging, we require wavelength-tunable burst sequences in the vicinity of 226 nm, corresponding to the NO single photon “\( \gamma \) band” \( A^2\Sigma^+ \rightarrow X^2\Pi \) electronic absorption. The fundamental wavelength of the pulse burst
laser is 1064 nm. To obtain a tunable wavelength at ~226nm, we employ Second Harmonic Generation and Third Harmonic Generation to obtain 355nm burst sequences. By using a widely tunable Optical Parametric Oscillator (OPO) system, which is pumped at 355nm, we can obtain 622nm sequences. Finally, by mixing the 622nm and 355nm outputs we can reach our target, wavelength tunable burst sequences at 226 nm. For comparison, we also attempted to use OPO output at 452 nm and obtain 226 nm by second harmonics generation. This work will be described in detail in Chapter 3.

1.3 High-repetition rate wavelength-tunable laser system

1.3.1 Dye laser and other systems

Planar Laser-Induced Fluorescence (PLIF) imaging of species such as CH, NO, and OH can yield considerable insight into the dynamics of unsteady and/or turbulent reacting flows. Since PLIF requires absorption of resonant radiation, it is necessary to utilize suitable wavelength-tunable laser sources. While dye and/or solid-state sources, including Nd:YAG-pumped OPOs, are readily available commercially, the published technology on generation of high-speed bursts of such tunable radiation is, to our knowledge, limited to a few papers.

Wu, et al. \cite{wu} pumped a grazing-incidence dye laser with ~0.5 mJ per pulse at 532 nm from an Nd:YAG burst-mode laser which is similar to that described in this work. While burst-mode output was obtained, the individual pulse energies were too low to be
measured. Because the pumping rate is 1 MHz, and liquid dye cannot circulate fast enough, only about half of the input 30 pulses generated observable output.

Luff, et al. [22] have reported an alexandrite system which utilizes a combination of a long pulse (~170 nsec) Q-switched oscillator, a Harriott Cell – Regenerative Amplifier (for pulse slicing and amplification), and two single pass amplifiers. Approximately 150 microjoules per pulse, for a 30-pulse sequence with 1 usec separation was reported. Each pulse is 20 nsec with a spectral line-width of ~ 0.1 nm at a wavelength of 761 nm. The output energy is too low to be used for generation of UV light.

Kaminski et al. [3] and Hult et al. [4] used the four commercial double-pulsed Nd:YAG lasers described previously to pump a single commercial dye laser. The system was used for OH PLIF imaging. Starting with 270 mJ per individual pulse at 532 nm, they were able to generate eight pulses at 282 nm, with an average energy of ~ 1 mJ and a minimum interpulse period of 125 µsec, constrained by the high intensity pumping requirement of the dye laser. They also found that the output energy and beam profile deteriorate for consecutive pulses in a sequence for the same reason. The energy decay can be compensated for by reduction of the pumping energy in the first pulses until all output pulses from the dye laser have the same energy. Unfortunately, this procedure strongly reduces the output energy in each pulse compared with the output energy from single-pulse pumping.

Based on the above, it was concluded that although dye lasers are an attractive solution for use with commercial, 10 Hz, pump lasers, they are not suitable for ultra-high repetition rate systems. This is due to the limited time available for the dye solution in the
dye cells to be exchanged between consecutive pumping pulses. For this reason, we chose to develop a new approach, based on the use of an Optical Parametric Oscillator (OPO) instead of the conventional dye laser.

1.3.2 OPO system

Optical Parametric Oscillator \(^{[23]}\) is a widely tunable optical generation system. OPO is in some respects the opposite of harmonic generation. Figure 1.1 shows a typical OPO cavity. If a nonlinear crystal is put in a cavity, the pump laser beam will be converted to two longer wavelength beams. This process is accomplished by the use of non-linear crystals. The two output beams are termed "signal" and "idler" waves, having a wavelength of \(\lambda_s\) and \(\lambda_i\), and the incident laser wave is termed "pump" wave with wavelength \(\lambda_p\). Under proper conditions, as will be described in more detail in Chapter 3, the idler wave can mix with the pump beam to produce a traveling polarization wave at the signal frequency, phased such that growth of the signal wave results. The process continues with the signal and idler waves both growing, and the pump wave decaying as a function of distance in the crystal.

![Figure 1.1: A typical OPO cavity](image-url)

M1: Reflecting \(\nu_s, \nu_i\), Transmitting \(\nu_p\)  
M2: Partially transmitting

Figure 1.1: A typical OPO cavity
Since each pump photon with energy $h\nu_p$ is generating a photon at the signal ($h\nu_s$) and idler frequency ($h\nu_i$), energy conservation requires that

$$\frac{1}{\lambda_p} = \frac{1}{\lambda_s} + \frac{1}{\lambda_i} \quad (1.1)$$

In order to achieve significant parametric amplification, it is required that at each of the three frequencies the generated polarization waves travel at the same velocity as a freely propagating electromagnetic wave. As will be shown, this will be the case if the refractive indices of the material are such that the $k$ vectors satisfy the momentum-matching condition

$$k_p = k_s + k_i \quad \text{where} \quad k_p = \frac{\omega_p n_p}{c} = \frac{2\pi n_p}{\lambda_p} \quad (1.2)$$

For collinearly propagating waves this may be written:

$$\frac{n(\lambda_p)}{\lambda_p} = \frac{n(\lambda_s)}{\lambda_s} + \frac{n(\lambda_i)}{\lambda_i} \quad (1.3)$$

where $\lambda_p$, $\lambda_s$, and $\lambda_i$ are the wavelengths of the pump, signal, and idler beams, respectively, and $n(\lambda)$ is the wavelength-dependent index-of-refraction. This is also called the Phase-matching condition.

Since the three indices of refraction depend on the wavelength, the direction of propagation in the crystal and on the polarization of the waves, it is generally possible by using birefringence and dispersion to find conditions under which the Phase-matching condition is satisfied.
Tunability is a fundamental characteristic of all parametric devices. With the pump providing input at the fixed wavelength $\lambda_p$, small changes of the refractive index around the phase matching condition, will change the signal and idler wavelengths such that a new phase-matching condition is achieved. Just like the second harmonic generation, tuning is possible by making use of the angular dependence of the birefringence of anisotropic crystals, and also by temperature variation. They are called Critical Phase Matching and Non-critical Phase Matching, respectively.

In Figure 1.1, M1 is a high reflectivity mirror, which reflects both signal and idler wavelengths, and M2 is a partial reflector. This cavity is termed a Doubly Resonant Cavity (DRO). If the high reflector M1 only reflects the signal wavelength, this kind of OPO system is termed a Singly Resonant Cavity (SRO). Typical OPOs have signal/idler tunability in the range 410 nm – 2600 nm for 355 nm pump, and 680 – 3000 nm for 532 nm pump.

The first successful operation of an OPO was achieved by Giordmaine and Miller in 1965\cite{24}. Since commercial 10 Hz laser pumped OPOs are readily available now, they have been used widely in many research areas, in flow diagnostics, in combustion diagnostics and even in disease diagnostics \cite{25}. The third harmonic output of Nd:YAG at 355 nm, is the most popular pump wavelength for OPO systems, because the output signal wavelength is shorter, $\sim$ 410 nm , than that achievable when pumped at 532 nm (680 nm).

We use the OPO process for high repetition rate generation of tunable radiation, in order to take advantage of the fact that the gain decays essentially instantaneously after
the passage of each pump pulse within the burst. To our knowledge, OPO has not been used for high repetition rate high-speed flow imaging, due to the inability to simultaneous generate high (~MHz) repetition rate and high (~mJ/pulse) pulse energies. It has, however, been well studied for use in optical spectroscopy serving as a high repetition rate, low energy (~µJ or nJ) wavelength-tunable source.

Phillips et al. [26] have reported a low-pump-threshold, all-solid-state subpicosecond OPO for the mid-infrared, based on Periodically Poled LiNbO$_3$ (PPLN) as the OPO gain medium, pumped by a mode-locked Ti:sapphire laser. Mid-infrared (3.9-6 µm) average idler power of 64 mW has been reported at pulse repetition rates of up to 322 MHz corresponding to ~0.2 nJ/pulse. The average signal output power is ~280 mW. The reported conversion efficiency is ~35% and the pulse width is ~ 0.4 ps.

Agnesi et al. [27] also pumped a PPLN OPO which is tunable in the 1.45 – 1.56 µm spectral range. They used a diode-pumped mode-locked Nd:YVO$_4$ laser as the pump source. The pulse width is adjustable from 1 ps – 15 ps with a 200 MHz repetition rate. With this OPO system they obtained 630 mW output power with 2.5 W input power.

Many other researchers have reported high repetition rate OPO system pumped by 100s MHz Nd:YVO$_4$ laser. As we stated previously, since the pump source does not operate in burst mode, high repetition rate constrains individual pulse energy to be too low to be used in flow imaging studies.

There were, however, a variety of issues which needed to be addressed in order to successfully achieve burst mode laser pumped OPO output, such as achieving narrow-linewidth, overcoming thermal effects, etc. The narrow-linewidth requirement makes the
phase-matching condition harder and thermal effect will result in lower conversion ratio, as will be discussed in Chapter 3.

1.4 NO PLIF

NO is a common tracer species for study of nonreacting flow because of its thermal stability, well-characterized spectroscopy, and strong absorption at 226 nm, corresponding to the $A^2\Sigma^+ - X^2\Pi$ “γ band” (0,0) transition. NO has been used as a tracer in flow diagnostics since the 1950s\textsuperscript{[28]}, and numerous studies has been performed to determine its molecular structure, spectroscopy of absorption and fluorescence, Einstein coefficients, etc. It is such an interesting molecule that some researchers have even studied and used it in the medical area to cure heart disease. (For which the Nobel Prize in Medicine was awarded in 1998.)

1.4.1 The Spectroscopy of the NO Molecule

A. Spectroscopy

Figure 1.2 shows an energy level diagram for NO and Fig. 1.3 shows the full potential curves\textsuperscript{[29]. Figure 1.2 indicates that the NO spectrum contains four electronic band systems, the $\beta$, $\gamma$, $\delta$, and $\epsilon$. Two of these band systems, the $\delta$ and $\epsilon$, are located in the vacuum-ultraviolet (below $\sim$ 180 nm), respectively, and the remaining two, the $\gamma$ and the $\beta$, in the near-ultraviolet and visible, thus making them especially important to fluorescence studies. The ground state of the NO molecule is $X^2\Pi$. This is also the
common lower level for the $\text{A}^2\Sigma^+ - \text{X}^2\Pi_{1/2, 3/2}$ band system. The $\text{X}^2\Pi$ ground state is a doublet with $\Delta E = 124 \text{ cm}^{-1}$, so that each of the NO bands has a characteristic double head.

Figure 1.2: Energy level diagram of the key electronic states for the NO molecule.

Figure 1.3: Potential curves for the key electronic states of the NO molecule.

Figure 1.4 is a schematic representation of the rotational terms and transitions for some of the first lines in the $\gamma$-band system of NO. The symbols $c$ and $d$, and $(+)$ and $(-)$, respectively, indicate the symmetric and antisymmetric, and the even and odd, components of the rotational lines resulting from $\lambda$ splitting. Spin-orbit coupling causes
the lower $^2$Π state to split into two levels, the $^2$Π$_{1/2}$ and the $^2$Π$_{3/2}$ and leads to the appearance of two band subsystems, each with six branches.

![Diagram](image)

Figure 1.4: A schematic representation of the rotational terms and transitions for some of the first lines in the $\gamma$-band system of NO.

Four of the six subordinate branches, namely the $P_{Q12}$, $Q_{R12}$, $Q_{P21}$, and $R_{Q21}$, completely merge with the corresponding lines of the principal $P_2$ $Q_2$, $Q_1$, and $R_1$ branches, thereby reducing the number of observed branches to eight. The presence of these subordinate branches does, however, affect line intensities where superposition occurs. The principal branches $P_2$ and $R_1$ have no subordinate branches. The subordinate branches $Q_{P12}$ and $S_{R12}$ result from transitions in which $\Delta K = 2$, $J$ being the total angular momentum of the molecule and $K$ the difference between the total angular momentum
and the spin. Such transitions would be completely forbidden in the Hund b case, where the K selection rules are strictly followed. It has already been pointed out that the $^2\Pi$-state coupling is usually intermediate between the Hund types $a$ and $b$. Here the coupling approximates type $a$ at low values of $J$ and type $b$ at high values of $J$.

The upper level for the NO $\gamma$-system shows a Hund type-b coupling, as is always true of $^2\Sigma$ states. The coupling in the lower $\chi^2\Pi$ state is intermediate between the Hund types $a$ and $b$. As a result, neither the $J$ nor the $K$ selection rules are strictly satisfied; this, in conjunction with doublet and $\Lambda$ splitting, gives rise to twelve branches, or sequences of rotational lines, which differ from one another in intensity.

B. Intensity Factors

The NO molecular constants can be found in reference [30]. Danielak et al. [31] reinvestigated the $\gamma$ band emission of the NO molecule and published a new version of NO molecular constants, Frank-Condon factors and rotational structure constants. Piper et al. [32] also reported the Einstein coefficients in detail for NO $\gamma$ band.

Earls[33] has developed simple expressions for the Henley-London factors, $S_{J', J''}$ of diatomic molecules, in $A^2\Sigma--X^2\Pi$ transitions with a type-b coupling of angular momentum in the $^2\Sigma$ level and a coupling intermediate between types $a$ and $b$ in the $^2\Pi$ level. This is exactly the case met in the $\gamma$ bands of NO.
The dependence of the intensity factor on the upper-level rotational quantum number is shown for the $\gamma$ system of NO in Fig. 1.5. It is seen that the largest $S_{\gamma}$ factors are associated with the $Q_1$ and $Q_2$ bands where $S^{Q_1} = S^{Q_2}$; the factors for the $P_1$, $P_2$, $R_1$ and $R_2$ branches are almost identical, and approximately only half as large as the $Q$-branch factors at high values of $J$.

Another intensity factor is the population distribution. The equilibrium expression is the Boltzmann Distribution. At room temperature $T=300K$, most of the NO molecules reside in the $v=0$ ground state. For the rotational population distribution, NO exhibits a maximum at $J=7$, at $T=300K$. 

Figure 1.5: The variation of the Henley-London factor with the rotational quantum number $J$ of the NO $\gamma$-band.
For the calculation of the absorption and fluorescence spectrum, we also need to consider the line broadening effect. There are Doppler Broadening, Pressure broadening and the combination effect Voigt Broadening, which can be found in detail in reference [34].

C. Spectrum

![Experimental setup for NO fluorescence spectrum measurement.](image)

Figure 1.6: Experimental setup for NO fluorescence spectrum measurement.

For better understanding of the fine structure of the NO absorption and fluorescence spectrum, we performed an initial set of spectral measurements. Figure 1.6 shows the experimental setup. A dye laser is pumped by the second harmonic output (532 nm) of a commercial 10 Hz repetition rate Nd:YAG laser, with output at 622nm. The third harmonic of the Nd:YAG laser (355 nm) mixes with the dye laser output in a Type I
BBO crystal to produce 226 nm light, which is used to excite NO molecules in a flow cell. We use a monochromator, photomultiplier tube, and boxcar integrator for detection, and PC controlled data acquisition system.

![Figure 1.7: NO absorption spectrum obtained by tuning the dye laser wavelength.](image)

Figure 1.7 displays the NO LIF excitation spectrum for a mixture consisting of 6 torr of NO and 16 torr N\(_2\), where the detection monochromator is fixed at a wavelength of 247 nm, which corresponds to \(A^2\Sigma(v'=0) \rightarrow X^2\Pi(v''=2)\) fluorescence emission. The monochromator has a detection bandwidth of ~±4 nm. In order to capture the rotational structure of the spectrum, we tune the dye laser slowly in 10\(^{-4}\) nm (~2.6×10\(^{-3}\) cm\(^{-1}\)) increments in the vicinity of 622 nm, which due to the mixing process, corresponds to a resolution of 10\(^{-5}\) nm at the NO absorption wavelength of 226 nm. The 226 nm laser power is ~1mJ/pulse. The purpose of this experiment was to search for the optimal wavelength region to excite NO for subsequent PLIF measurements (to be described in
Chapter 4), so we didn’t measure the fluorescence intensity carefully. Specifically, the fluorescence shown in Figure 1.7 maybe exhibit some partial saturation, so that the relative intensity may be somewhat in error. However, the NO absorption wavelengths are not affected. The result shown in Figure 1.7 confirms that NO has a very wide absorption range at ~ 226 nm at room temperature, with a multitude of individual transitions.

![Fluorescence Spectrum](image)

**Figure 1.8:** NO dispersed fluorescence spectrum.

Figure 1.8 shows the dispersed fluorescence spectrum with the same experimental conditions as used in Figure 1.7. In this case, we fix the UV excitation wavelength at ~226 nm and scan the detection spectrometer in 1nm steps to measure the fluorescence
spectrum. There are seven peaks in Figure 1.8, which correspond to NO γ band $0\rightarrow0$, $0\rightarrow1$, $0\rightarrow2$, $0\rightarrow3$, $0\rightarrow4$, $0\rightarrow5$ and $0\rightarrow6$ vibrational transitions, respectively from left to right. (Note that Rayleigh/Mie scattering is very weak and can be neglected.) Figure 1.8 tells us that NO has very strong fluorescence in the wavelength range 220-300nm. However, to obtain the NO PLIF images, we need an imaging camera with high sensitivity in this wavelength range.

1.4.2 Planar Laser Induced Fluorescence

Planar Laser Induced Fluorescence (PLIF) imaging of NO has been well studied for many years, both in flow visualization and in combustion. Normally, people use the frequency-doubled output of an Nd:YAG laser or excimer laser pumped dye laser to obtain 226 nm laser pulses. For this work we use an OPO instead of the dye laser as a source of tunable radiation. The fluorescence lifetime $^{[32]}$ of NO is $\sim$200ns. Some basic LIF formulas $^{[35]}$ describing the LIF process are as follows:

The saturation spectral irradiance is defined as

$$I_{v}^{sat} = \frac{(A_{21} + Q_{21})c}{B_{12} + B_{21}}$$  \hspace{1cm} (1.4)$$

where $A_{21}$, $Q_{21}$ and $B_{12}$ are rate constants for spontaneous emission, quenching and stimulated absorption, respectively. The relation between Einstein A and B coefficients is given by:
\[ \frac{A_{21}}{B_{12}} = \frac{g_1}{g_2} \frac{8\pi n^3 \hbar}{\lambda^3} \]  

(1.5)

where \( g_1 \) and \( g_2 \) are the degeneracy.

The detected fluorescence signal power is expressed as

\[ F = h\nu \frac{d\Omega}{4\pi} lAN \frac{A_{21}}{1 + \frac{I_{\text{sat}}}{I_v}} \]  

(1.6)

where \( d\Omega \) is the collection solid angle (in steradians); \( A \) is the cross-sectional area of the laser beam; \( N \), the number density of molecules on the rotation-vibrational level being excited; and \( l \), the path length for which the fluorescence is observed. Note that for \( I \ll I_{\text{sat}} \), the detected signal, \( F \), is linear with laser intensity. However, as \( I_v \) approaches \( I_{\text{sat}} \), the signal increases less than linearly, eventually becoming independent of \( I_v \), at very high intensity.

1.4.3 A Brief History of NO PLIF Research

To our knowledge, the first NO based laser induced fluorescence work was reported by Grieser and Barnes \[36\] in 1980. They used laser induced fluorescence to detect nitric oxide in a CH\(_4\)-O\(_2\)-N\(_2\) flame at atmospheric pressure. A frequency-doubled tunable dye laser, pumped by a pulsed-nitrogen laser, was used to excite rotational transitions in the (0,0) \( \gamma \) band of NO. The laser pulse repetition rate was 30 Hz.
Seitzman et al.\textsuperscript{[37]} described a single-pulse, PLIF diagnostic for the measurement of two-dimensional temperature fields in combustion flows. They used a frequency-doubled Nd:YAG laser to pump a pulsed dye laser. The doubled output of the dye laser was summed with the residual Nd:YAG 1064 nm beam in a KD\textsuperscript{+}P crystal to obtain output (~0.5 mJ/pulse) at a wavelength of 225.6 nm corresponding to the Q1(22) line of the NO $\gamma$ band transition. They made a 2-D temperature measurement in a methane-air flame using the NO PLIF method.

McMillin et al.\textsuperscript{[38]} used the frequency-doubled output XeCl excimer-pumped dye laser as the excitation source. They made NO PLIF measurements in a Scramjet model flowfield with a two-line rotational PLIF technique. Their laser pulse energy is $\sim$ 0.2-0.4 mJ. They showed NO temperature images with a temperature range from 250K to 2000K.

Rossmann et al.\textsuperscript{[39]} performed NO PLIF imaging technique in a low-pressure hypersonic mixing flow, with a convective Mach number of 2.64, and a Mach 4.87 flow over a 21\textdegree, two-dimensional wedge, driven by a shock tunnel. They used the frequency-tripled output of a Nd:YAG laser (355nm, 280 mJ) to pump a blue dye. The dye-laser output (452 nm, 30 mJ) is doubled with a BBO crystal. They obtained UV pulses at 226 nm with 0.2 mJ, 6.8 ns duration, and $\sim$0.5 cm\textsuperscript{-1} spectral width.

Bessler et al.\textsuperscript{[40]} performed NO PLIF measurements in premixed methane and air flames at high-pressure flames, with a pressure range between 1 and 60 bars. They also used the frequency-doubled output of a blue dye laser, which is pumped by the Third Harmonic output of an Nd:YAG laser. They studied several excitation wavelengths for different temperature conditions of the flame. At low temperature, they selected 226.87
nm, and whereas at high temperature, they selected 224.82 nm as the excitation wavelengths.

Meyer et al. [41] used NO PLIF and Acetone PLIF simultaneously to study molecular mixing in high-speed gaseous flows. They generated the NO excitation wavelength of 226 nm with the same method as reference [39]. NO was seeded into the core-flow and acetone into the co-flow air. By comparing NO PLIF images with the acetone PLIF images, they obtained images of mixed jet fluid fraction.

These studies represent just a few drops in the sea of NO PLIF research. Nonetheless they serve to illustrate the wide use of NO PLIF in flow and combustion studies over a wide temperature and pressure range. We note that while the tunable dye laser has been used almost exclusively in these studies, it is not suitable for ultra-high repetition rate imaging, for the reasons noted previously.
CHAPTER 2

INTRODUCTION TO NONLINEAR OPTICS

In this chapter we provide an introduction and background to nonlinear optics \cite{42}. The nonlinear effect can give rise to the exchange of energy between a number of electromagnetic fields of different frequencies. Three of the most important applications of this nonlinear phenomenon are (1) second harmonic generation, in which part of the energy of an optical wave of frequency $\omega$ propagating through a crystal is converted to that of a wave at $2\omega$; (2) optical parametric oscillation, in which a strong pump wave at $\omega_3$ causes the simultaneous generation in a nonlinear crystal, of radiation at $\omega_1$ and $\omega_2$, where $\omega_3 = \omega_1 + \omega_2$; (3) frequency up-conversion, including third harmonic generation and electromagnetic fields mixing, in which a weak signal of a low frequency $\omega_1$ is converted coherently to a signal of a higher frequency $\omega_3$ by mixing with another electromagnetic field at $\omega_2 = \omega_3 - \omega_1$. 
2.1 Introduction of Electromagnetic Fields

A. Maxwell’s equations

This section provides the main background for classical electromagnetic theory. The starting point is Maxwell’s equations.

\[ \nabla \times \vec{H} = \vec{i} + \frac{\partial \vec{D}}{\partial t} \quad (2.1) \]
\[ \nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad (2.2) \]

and the constitutive equations relating the polarization of the medium to the displacement vectors

\[ \vec{D} = \varepsilon_0 \vec{E} + \vec{P} \quad (2.3) \]
\[ \vec{B} = \mu_0 (\vec{H} + \vec{M}) \quad (2.4) \]

where \( \vec{i} \) is the current density (amperes per square meter); \( \vec{E}(\vec{r},t) \) and \( \vec{H}(\vec{r},t) \) are the electric and magnetic field vectors, respectively; \( \vec{D}(\vec{r},t) \) and \( \vec{B}(\vec{r},t) \) are the electric and magnetic displacement vectors; \( \vec{P}(\vec{r},t) \) and \( \vec{M}(\vec{r},t) \) are the electric and magnetic polarizations (dipole moment per unit volume) of the medium; and \( \varepsilon_0 \) and \( \mu_0 \) are the electric and magnetic permeabilities of vacuum, respectively. A detailed discussion of Maxwell’s equation can be found in any standard textbook on electromagnetic theory, such as reference [43].
B. Power transport and storage

From Maxwell’s equations, it is easy to obtain an expression of the total power flowing into the volume $V$ bounded by a surface $S$

\[-\int_V \nabla \cdot (E \times H)\,dv \]
\[= \int_S (E \times H) \cdot \mathbf{n}\,ds \]

\[= \int_V \left[ E \cdot \frac{\partial}{\partial t} \left( \frac{\varepsilon_0}{2} E \cdot E \right) + \frac{\partial}{\partial t} \left( \frac{\mu_0}{2} H \cdot H \right) + \mu_0 \frac{\partial M}{\partial t} \right] \,dv \]

(2.5)

The first term on the right side is the power expended by the field on the moving charges; the sum of the second and third terms corresponds to the rate of increase of the vacuum electromagnetic stored energy; the next-to-last term represents the power per unit volume expended by the field on “driving” the electric dipoles, and the last term is the power expended by the field on the magnetic dipoles.

C. Dipolar dissipation in harmonic fields

According to the discussion in the last section, the average power per unit volume expended by the field on sustaining an electric polarization of the medium is

\[
\frac{\text{Power}}{\text{Volume}} = \overline{E \cdot \frac{\partial P}{\partial t}}
\]

(2.6)

where the horizontal bar donates time averaging. Let us assume that $\overline{E(t)}$ and $\overline{P(t)}$ are parallel to each other as

\[
E(t) = \text{Re}(Ee^{i\omega t}) ; \quad P(t) = \text{Re}(Pe^{i\omega t})
\]

(2.7)
where $E$ and $P$ are the complex amplitudes. The linear electric susceptibility $\chi_e$ is defined by

$$ P = \varepsilon_0 \chi_e E $$  \hspace{1cm} (2.8)

Then, combined equations (2.6), (2.7) and (2.8), we can obtain the following expression

$$ \frac{\text{Power}}{\text{Volume}} = \frac{\omega}{2} \varepsilon_0 |E|^2 \text{Re}(i\chi_e) $$  \hspace{1cm} (2.9)

In anisotropic media, equation (2.8) and (2.9) will be changed to

$$ P_i = \varepsilon_0 \sum_j \chi_{ij} E_j ; $$

and

$$ \frac{\text{Power}}{\text{Volume}} = \frac{\omega}{2} \varepsilon_0 \sum_{i,j} \text{Re}(i\chi_{ij} E_i^* E_j) $$  \hspace{1cm} (2.10)

where the asterisk superscript stands for the complex conjugate.

D. Propagation of light in anisotropic crystals

An understanding of wave propagation in anisotropic crystals is a prerequisite to the important topic, phase matching in nonlinear optical interactions. In an anisotropic crystal, the polarization induced by an electric field and the field itself, are not necessarily parallel. The electric displacement vector $\vec{D}$ and the electric field $\vec{E}$ are consequently related by means of the dielectric tensor $\varepsilon_{kl}$ defined by

$$ D_k = \varepsilon_{kl} E_l $$  \hspace{1cm} (2.11)

where the subscripts refer to a Cartesian coordinate ($k, l=x, y, z$).
In an isotropic medium, the stored electric energy density is given by

$$\rho_e = \frac{1}{2} \overrightarrow{E} \cdot \overrightarrow{D} = \frac{1}{2} E_k \varepsilon_{kl} E_l = \frac{1}{2} \varepsilon |E|^2$$  \hspace{1cm} (2.12)$$

Then in an anisotropic medium, the electric energy density $\rho_e$ can be written as

$$2 \rho_e = \varepsilon_{xx} E_x^2 + \varepsilon_{yy} E_y^2 + \varepsilon_{zz} E_z^2 + 2 \varepsilon_{xy} E_x E_y + 2 \varepsilon_{yz} E_y E_z + 2 \varepsilon_{xz} E_x E_z$$  \hspace{1cm} (2.13)$$

We can define a new Cartesian coordinate system to diagonalize (2.13). With the new system, $\rho_e$ becomes

$$2 \rho_e = \varepsilon_x E_x^2 + \varepsilon_y E_y^2 + \varepsilon_z E_z^2$$  \hspace{1cm} (2.14)$$

The new coordinate axes are called the principal dielectric axes. In the principal dielectric coordinate system, the tensor $\varepsilon_{kl}$ is diagonal and is given by

$$\begin{pmatrix} D_x \\ D_y \\ D_z \end{pmatrix} = \begin{pmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$  \hspace{1cm} (2.15)$$

Using (2.14) and (2.15), we obtain

$$2 \rho_e = \frac{D_x^2}{\varepsilon_x} + \frac{D_y^2}{\varepsilon_y} + \frac{D_z^2}{\varepsilon_z}$$  \hspace{1cm} (2.16)$$

If we use the definition of $\overrightarrow{D}$ and index of refraction $n$

$$n_k^2 = \frac{\varepsilon_k}{\varepsilon_0}, \ D = x\overrightarrow{\sqrt{2\rho_e\varepsilon_0}}$$  \hspace{1cm} (2.17)$$

Equation (2.16) can be written as

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1$$  \hspace{1cm} (2.18)$$
This is the equation of a general ellipsoid with major axis parallel to the \( x \), \( y \), and \( z \) directions whose respective lengths are \( 2n_x \), \( 2n_y \), and \( 2n_z \). The ellipsoid is known as the index ellipsoid.

In uniaxial crystals, that is, crystals in which the highest degree of rotational symmetry applies to no more than a single axis, the equation of the index ellipsoid simplifies to

\[
\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1
\]

(2.19)

where the axis of symmetry was chosen as the \( z \) axis. It is also referred to as the optical axis, \( n_o \) is called the ordinary index of refraction, whereas \( n_e \) is the extraordinary one. If \( n_e < n_o \), we have a negative (optically) uniaxial crystal, whereas in a positive crystal, \( n_e > n_o \).

We conclude that, given a specific propagation direction in a crystal, in general two possible linearly polarized modes exist, the “ordinary” ray and “extraordinary” ray of propagation. Each mode possesses a unique direction of polarization and a corresponding index of refraction. The existence of an “ordinary” and an “extraordinary” ray with different indices of refraction is called birefringence.

2.2 Introduction to Nonlinear Optics

We have previously considered the propagation of light in a linear medium in which the polarization is proportional to the electric field that induces it. In this section, we consider some of the consequences of the nonlinear dielectric properties of certain
classes of crystals in which polarization is produced that is proportional to the square of
the incident field. The first experiment in this nonlinear field took place in 1961 and
involved doubling of the frequency of a ruby laser in a quartz crystal \[44\].

A. The Nonlinear Optical Susceptibility Tensor

Consider the nonlinear coupling of two optical fields. The first, with frequency \(\omega_1\) and electric field polarization axis along the \(j\) direction, is given by

\[
E^{\omega_1}_j(t) = \text{Re}(E^{\omega_1}_j e^{i\omega_1 t}) = \frac{1}{2}(E^{\omega_1}_j e^{i\omega_1 t} + \text{c.c.})
\]

(2.20)

whereas the second field at \(\omega_2\) and polarization axis \(k\) is

\[
E^{\omega_2}_k(t) = \text{Re}(E^{\omega_2}_k e^{i\omega_2 t})
\]

(2.21)

If the medium is nonlinear, then the presence of these field components can give
rise to polarizations at frequencies \(n\omega_1 + m\omega_2\) where \(n\) and \(m\) are any integers. If we take
the polarization component at \(\omega_3 = \omega_1 + \omega_2\) along the \(I\) direction as

\[
P^{\omega_3}_i(t) = \text{Re}(P^{\omega_3}_i e^{i\omega_3 t})
\]

(2.22)

the nonlinear susceptibility tensor \(d^{\omega_3 = \omega_1 + \omega_2}_{y_k}\) is defined by the following relations

between the complex field amplitudes:

\[
P^{\omega_3}_i = 2d^{\omega_3}_{y_k} E^{\omega_1}_j E^{\omega_2}_k
\]

(2.23)

where we sum over repeated indices. (Note that some books define \(\varepsilon_d d\) instead of
\(d\) here as the nonlinear coefficient. In that case, \(d\) has a unit of \(m/V\).)
Similarly, we can define the difference frequency susceptibility tensor \( d_{ijk}^{\omega_3-\omega_1-\omega_2} \) by

\[
P_i^{\omega_1} = 2d_{ijk}^{\omega_1} E_j^{\omega_2} E_k^{\omega_2-\omega_1}
\]  
(2.24)

where, according to (2.20), \( E_k^{\omega_2-\omega_1} = (E_k^{\omega_2})^* \).

In most nonlinear frequency generation techniques, the crystal employed is transparent over a region that includes \( \omega_1 \), \( \omega_2 \), and \( \omega_3 \). This implies that \( d_{ijk} \) is independent of frequency. We can consequently express the nonlinear polarization as

\[
P_i(t) = 2d_{ijk} E_j(t) E_k(t)
\]  
(2.25)

\( d_{ijk} \) is also called the nonlinear coefficient.

B. On the Physical Origins of the Nonlinear Coefficient

Bloembergen \[45\] has developed a popular model of an anharmonic oscillator to discuss the nonlinear optical susceptibility. The same model was used by Garrett and Robinson \[46\] to derive an expression for the one-dimensional nonlinear coefficient.

The model assumes that the electronic response to a driving electric field can be simulated by that of an electron in an anharmonic potential well. The equation of motion for the electron is then

\[
\dot{X} + \gamma \dot{X} + \omega_0^2 X + DX^2 = \frac{eE_0}{2m} (e^{-i\omega t} + e^{i\omega t})
\]  
(2.26)

where \( X \) is the deviation from the potential minimum, \( mDX^2 \) is the anharmonic restoring force [corresponding to the term \( mDX^3 / 3 \) in the potential], the driving electric
field is $E_0 \cos \omega t$, and $\gamma$ is the damping term. A detailed discussion of equation (2.26) can be found in reference [45, 46].

C. The Electromagnetic Formulation of the Nonlinear Interaction

In this part we will give some basic formulations of the nonlinear interaction, following closely the discussion presented in reference [42]. We start with Maxwell’s equations in a form which includes the polarization $P$ explicitly

$$\nabla \times \mathbf{H} = i + \frac{\partial \mathbf{D}}{\partial t} = i + \frac{\partial}{\partial t} (\varepsilon_0 \mathbf{E} + \mathbf{P})$$

(2.27)

$$\nabla \times \mathbf{E} = -\frac{\partial}{\partial t} (\mu_0 \mathbf{H})$$

(2.28)

The polarization $\mathbf{P}$ is made up of a linear and a nonlinear term

$$\mathbf{P} = \varepsilon_0 \chi_L \mathbf{E} + \mathbf{P}_{NL}$$

(2.29)

where:

$$(P_{NL})_i = 2\varepsilon_{ijk} E_j E_k$$

(2.30)

If we use the conductivity $\sigma$ (where $i = \sigma \mathbf{E}$) and $\varepsilon$ (where $\varepsilon = \varepsilon_0 (1 + \chi_L)$) in equation (2.27) and (2.28), we get

$$\nabla^2 \mathbf{E} = \mu_0 \sigma \frac{\partial \mathbf{E}}{\partial t} + \varepsilon_0 \varepsilon \frac{\partial^2 \mathbf{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}$$

(2.31)

At this point, we specialize the problem to one dimension by taking $\frac{\partial}{\partial x} = \frac{\partial}{\partial y} = 0$

and denoting the arbitrary direction of propagation as $z$. We also limit the consideration
to three frequencies $\omega_1$, $\omega_2$, and $\omega_3$ and take the corresponding fields to be in the form of traveling plane waves

$$E_i^{\omega_1}(z,t) = \frac{1}{2} \left[ E_{ii}(z) e^{i(\omega_1 t - k_1 z)} + c.c. \right]$$

$$E_k^{\omega_2}(z,t) = \frac{1}{2} \left[ E_{2k}(z) e^{i(\omega_2 t - k_2 z)} + c.c. \right]$$

(2.32)

$$E_j^{\omega_3}(z,t) = \frac{1}{2} \left[ E_{3j}(z) e^{i(\omega_3 t - k_3 z)} + c.c. \right]$$

where $i, k, j$ refer to the $E_1, E_2, E_3$, the polarization direction in Cartesian coordinates and can each take on values $x$ and $y$.

The $i$ component of the nonlinear polarization at $\omega_j = \omega_3 - \omega_2$, as an example, is given according to (2.30) and (2.32) as

$$\left( P_{NL}^{\omega_j}(z,t) \right)_i = d'_{ijk} E_{3j}(z) E_{2k}^*(z) e^{i(\omega_3 t - \omega_2 t - (k_3 - k_2)z)} + c.c. \tag{2.33}$$

Note that $d'_{ijk}$ is the $d_{\alpha\beta\gamma}$ tensor of (2.30) transformed from the crystal coordinate system to that used here to describe the field propagation [42].

Substituting (2.32) into the wave equation (2.31), and using the relation

$$k = \omega \sqrt{\mu_0 \varepsilon}$$

we obtain the solution

$$\frac{dE_{1i}}{dz} = -\frac{\sigma_1}{2} \sqrt{\frac{\mu_0}{\varepsilon_1}} E_{1i} - i\omega_1 \frac{\mu_0}{\varepsilon_1} d'_{ijk} E_{3j} E_{2k}^* e^{-i(k_3 - k_2 - k_1)z}$$

$$\frac{dE_{2k}}{dz} = -\frac{\sigma_2}{2} \sqrt{\frac{\mu_0}{\varepsilon_2}} E_{2k}^* + i\omega_2 \frac{\mu_0}{\varepsilon_2} d'_{kij} E_{1i} E_{3j}^* e^{-i(k_1 - k_3 + k_2)z} \tag{2.34}$$
\[
\frac{dE_{3j}}{dz} = -\frac{\sigma_3}{2} \sqrt{\frac{\mu_0}{\varepsilon_3}} E_{3j} - i\omega_3 \sqrt{\frac{\mu_0}{\varepsilon_3}} \varepsilon_{jk} \varepsilon_{kl} E_{3k} e^{-i(k_j + k_k - k_3)z}
\]

Equation (2.34) describes the growth (or attenuation) of the three electric fields as they propagate in the \(z\) direction.

Now we define the new field variables \(A_l\) by

\[
A_l = \sqrt{n_\omega} \frac{1}{\omega_l} E_l, \quad l=1, 2, 3
\]  

(2.35)

so that the intensity at \(\omega_l\) is

\[
I_l = \frac{P_l}{A_l} = \frac{1}{2} \frac{\varepsilon_{\omega l}}{\mu_0} |E_l|^2 = \frac{1}{2} \frac{\varepsilon_{\omega l}}{\mu_0} |A_l|^2
\]  

(2.36)

Since a photon’s energy is \(\hbar \omega_l\), it follows, from (2.36), that \(|A_l|^2\) is proportional to the photon flux at \(\omega_l\), the proportionality constant being independent of frequency.

Equation (2.34) can now be written as

\[
\frac{dA_1}{dz} = -\frac{1}{2} \alpha_1 A_1 - i\kappa A_1^* A_3 e^{-i(\Delta k)z}
\]

\[
\frac{dA_2^*}{dz} = -\frac{1}{2} \alpha_2 A_2^* + i\kappa A_1 A_2 e^{i(\Delta k)z}
\]  

(2.37)

\[
\frac{dA_3}{dz} = -\frac{1}{2} \alpha_3 A_3 - i\kappa A_1 A_2 e^{i(\Delta k)z}
\]

where the indices 1, 2, 3 are now used to label the polarization directions of the three fields \(\overline{E}_1, \overline{E}_2, \overline{E}_3\).

\[
\Delta k = k_3 - k_1 - k_2
\]
\[ \kappa = d_{123} \sqrt{\frac{\mu_0 \omega_1 \omega_2 \omega_3}{\varepsilon_0 n_1 n_2 n_3}} \]  

(2.38)

\[ \alpha_i = \sigma_i \sqrt{\frac{\mu_0}{\varepsilon_i}}, \quad i=1, 2, 3 \]

Equations (2.34) and (2.37) are the basic formulations of the nonlinear interaction. We will apply them to some specific cases in the following sections.

2.3 Second Harmonic Generation

A. Low Conversion Efficiency

Consider (2.34) for second harmonic generation (SHG). In SHG, \( \omega_1 = \omega_2 \), and \( \omega_3 = 2\omega_1 \). If we assume the total amount of power lost by the input beam is small so that

\[
\frac{dE_{1i}}{dz} \approx 0,
\]

we need to consider only the last equation in (2.34). If the medium is non-conducting at \( \omega_3 \), then \( \sigma_3 = 0 \), and we have

\[
\frac{dE_{3j}}{dz} = -2i\omega \sqrt{\frac{\mu_0}{\varepsilon}} d_{jik} E_{1i} E_{1k} e^{i\Delta k} \]  

(2.39)

where \( \omega = \omega_1 = \frac{\omega_3}{2} \), and \( \Delta k = k_j^{(i)} - k_j^{(i)} - k_j^{(k)} \)  

(2.40)

and where \( k_j^{(i)} \) is the propagation constant for the beam at \( \omega_i \), which is polarized along the \( i \) direction. The solution of (2.39) for \( E_{3j}(0) = 0 \) (i.e., no second harmonic input) and for a crystal of length \( L \) is
\[ E_{3j}(L) = -2i\omega \left( \frac{\mu_0}{\epsilon} d'_{jik} E_{1j} E_{1k} e^{i\Delta k L} - 1 \right) \]  \hspace{1cm} (2.41)

\[ E_{3j}(L)E^*_{3j}(L) = \frac{4}{\epsilon_3} \frac{\omega^2 (d'_{jik})^2 E_{1j}^2 E_{1k}^2 L^2}{\Delta k L / 2} \sin^2(\Delta k L / 2) \] \hspace{1cm} (2.42)

To obtain an expression for the second harmonic power output \( P^{(2\omega)} \), we use the relation

\[ I^{(2\omega)} = \frac{P^{(2\omega)}}{Area} = \frac{1}{2} \sqrt{\frac{\epsilon}{\mu_0}} E_{3j} E^*_{3j} \] \hspace{1cm} (2.43)

Using this expression, we obtain

\[ I^{(2\omega)} = 2 \sqrt{\frac{\mu_0}{\epsilon_3}} \omega^2 (d'_{jik})^2 E_{1j}^2 E_{1k}^2 L^2 \sin^2(\Delta k L / 2) \] \hspace{1cm} (2.44)

The conversion efficiency is thus

\[ \frac{P^{(2\omega)}}{P^{(\omega)}} = 8 \left( \frac{\mu_0}{\epsilon_0} \right)^{3/2} \frac{\omega^2 d'_{jik} L^2}{n^3} I^{(\omega)} \sin^2(\Delta k L / 2) \] \hspace{1cm} (2.45)

Here we use \( \epsilon_1 \approx \epsilon_3 = \epsilon_0 n^2 \)

B. General Second Harmonic Generation

We consider equations (2.37) for general second harmonic generation, so \( A_1 \) and \( A_2 \) are equal. (2.37) becomes

\[ \frac{dA_1}{dz} = -i\kappa A_1^* A_2 e^{-i(\Delta k)z} \] \hspace{1cm} and \hspace{1cm} \[ \frac{dA_2}{dz} = -i\kappa A_2^* A_1 e^{i(\Delta k)z} \] \hspace{1cm} (2.46)

If we choose \( A_1(0) \) as a real number and assume the phase matching condition, \( \Delta k = 0 \), the solution of (2.46) \[^{47}\] is
\[ A_j(z) = -iA_j(0) \tanh[\kappa A_j(0)z] \] 

(2.47)

The conversion efficiency is

\[ \frac{P^{(2\omega)}}{P^{(\omega)}} = \frac{|A_j(z)|^2}{|A_j(0)|^2} = \tanh^2[\kappa A_j(0)z] \] 

(2.48)

where \(\kappa\) is defined by (2.38).

2.4 Optical Parametric Amplification and Oscillation

Optical parametric amplification and oscillation in the simplest form involves the transfer of power from a “pump” wave at \(\omega_3\) to waves at frequencies \(\omega_1\) and \(\omega_2\), where \(\omega_3 = \omega_1 + \omega_2\). It is fundamentally similar to the case of second harmonic generation. The only difference is in the direction of power flow. In second harmonic generation, power is fed from the low-frequency optical field at \(\omega\) to the field at \(2\omega\). In parametric amplification, power flow is from the high-frequency field \(\omega_3\) to the low frequency fields at \(\omega_1\) and \(\omega_2\). In the special case where \(\omega_1 = \omega_2\), we have the exact reverse of second harmonic generation. This is the case of the so-called degenerate parametric amplification.

A. Basic Equations of Parametric Amplification

With the low conversion efficiency assumption, we take \(A_j(z) = A_j(0)\) in (2.37) and write the first two equations as
\[
\frac{dA_1}{dz} = -\frac{1}{2}\alpha_1 A_1 - igA_2^* e^{-i(2\Delta k)z}
\]

\[
\frac{dA_2^*}{dz} = -\frac{1}{2}\alpha_2 A_2^* + igA_1 e^{i(2\Delta k)z}
\]

(2.49)

where

\[2\Delta k = k_3 - k_1 - k_2\]

\[g = \kappa A_3(0) = \sqrt{\frac{\mu_0}{\varepsilon_0}} \frac{\omega_1 \omega_2}{n_1 n_2} d E_3(0)\]

(2.50)

Let us consider the general case in which both “signal” and “idler” waves with amplitudes \(A_1(0)\) and \(A_2(0)\), respectively, are present at the input. We also consider for simplicity the phase matching case \(\Delta k = 0\) in (2.49). The solution with \(\alpha_i = 0\) (no losses) is

\[A_1(z)e^{i(\Delta k)z} = A_1(0) \cosh(\gamma z) - iA_2^*(0) \sinh(\gamma z)\]

(2.51)

\[A_2^*(z)e^{-i(\Delta k)z} = A_2^*(0) \cosh(\gamma z) + iA_1(0) \sinh(\gamma z)\]

(2.52)

B. Parametric Oscillation

In the last section it was shown that a pump wave can provide, via interaction in a nonlinear crystal, simultaneous amplification for “signal” and “idler” waves. If the nonlinear crystal is placed within an optical resonator that provides resonances for the signal or idler waves, the parametric gain will, at some threshold pumping intensity, cause simultaneous oscillation at both the signal and idler frequencies. The threshold for this oscillation corresponds to the point at which the parametric gain just balances the
losses of the signal and idler waves. This is the physical basis of the optical parametric oscillator [48].

1) Doubly Resonant OPO (DRO)

Let us consider a DRO system, as shown in Figure 1.1. The high reflector M1 reflects both signal and idler wavelengths, and the output coupler M2 has reflectance of $R_1$ and $R_2$ at frequency $\omega_1$ and $\omega_2$, respectively. The threshold condition for the doubly resonant OPO is given by the following equation. A detailed discussion can be found in reference [42].

\[
(R_1 + R_2) \cosh(g_i l) - R_1 R_2 = 1
\]

(2.53)

where $l$ is the length of the nonlinear crystal and $g_i$ is defined by (2.50).

If we express the pump field $E_p$ in terms of the intensity, which is defined by (2.36) in equation (2.50), equation (2.53) becomes

\[
I_{3t} = \left( \frac{\varepsilon_0}{\mu_0} \right)^2 n_1 n_2 n_3 \left( \cosh^{-1} \frac{1 + R_1 R_2}{R_1 + R_2} \right)^2 \frac{8 \omega_1 \omega_2 l^2 d^2}{\varepsilon_0}
\]

(2.54)

2) Singly Resonant OPO (SRO)

In the singly resonant OPO, only one frequency oscillates in the cavity, so $R_2 = 0$ in (2.53). In this case, (2.53) becomes

\[
R_1 \cosh(g_i l) = 1
\]

(2.55)
The threshold pump intensity is

\[
I_{3t} = \left( \frac{\varepsilon_0}{\mu_0} \right)^{3/2} \frac{n_1 n_2 n_3}{8 \omega_1 \omega_2 \mu_1 l^2} \left( \coth \left( \frac{1}{R_I} \right) \right)^2
\]

(2.56)

2.5 Frequency Up-conversion

Parametric interactions in a nonlinear crystal can be used to convert a signal from a low frequency \( \omega_i \) to a high frequency \( \omega_j \) by mixing it with a strong laser beam at \( \omega_2 \), where \( \omega_i + \omega_2 = \omega_j \). We can consider frequency up-conversion to be a general expression of second harmonic generation.

The analysis of frequency up-conversion starts with (2.37). Assuming negligible depletion of the strong laser field \( A_j \), no losses (\( \alpha = 0 \)) at \( \omega_i \) and \( \omega_j \) and phase matching (\( \Delta k = 0 \)), we can write (2.37) as

\[
\frac{dA_i}{dz} = -igA_3
\]

\[
\frac{dA_j}{dz} = -igA_i
\]

(2.57)

where if we use (2.35) and (2.38) and choose without loss of generality the phase of \( A_j \) as zero so that \( A_j(0) = A_j^*(0) \),

\[
g = \sqrt{\frac{\omega_j \omega_3}{n_1 n_3} \left( \frac{\mu_0}{\varepsilon_0} \right)} dE_2
\]

(2.58)
The general solution of (2.57) is

\[ A_i(z) = A_i(0) \cos(gz) - iA_i(0) \sin(gz) \]

\[ A_j(z) = A_j(0) \cos(gz) - iA_j(0) \sin(gz) \] (2.59)

If the input frequencies are only \( \omega_i \) and \( \omega_j \), we have \( A_j(0) = 0 \). In this case,

\[ |A_i(z)|^2 = |A_i(0)|^2 \cos^2(gz) \]

\[ |A_j(z)|^2 = |A_j(0)|^2 \sin^2(gz) \] (2.60)

If we use the intensity expression in (2.36), we can rewrite (2.60) in terms of powers.

\[ P_i(z) = P_i(0) \cos^2(gz) \]

\[ P_j(z) = \frac{\omega_j}{\omega_i} P_j(0) \sin^2(gz) \] (2.61)

In a crystal of length \( l \), the conversion efficiency is thus

\[ \frac{P_3(l)}{P_1(0)} = \frac{\omega_3}{\omega_1} \sin^2(gl) \] (2.62)

Using (2.36) and (2.58), we can rewrite equation (2.62) as

\[ \frac{P_3(l)}{P_1(0)} = \frac{\omega_i}{\omega_1} \sin^2 \left( dl \sqrt{\frac{2I_3 \omega_i \omega_3}{n_2 n_3}} \left( \frac{\mu_0}{\varepsilon_0} \right)^{3/2} \right) \] (2.63)
CHAPTER 3

DEVELOPMENT OF THE MHZ REPETITION RATE PULSE BURST LASER

The pulse burst laser is an Nd:YAG based system capable of providing a burst of up to 99 short duration (~10 nsec) pulses at repetition rates up to 1 MHz. The system is shown schematically in Figure 3.1. A continuous wave Nd:YAG ring laser (Lightwave Electronics Series 126) serves as the primary oscillator. It is a narrow linewidth (<5 kHz), single frequency laser which can be tuned over a 35 GHz range by adjusting the temperature of the laser crystal. The output is 100 mW at 1.064 microns and is subsequently pre-amplified in a double-pass flashlamp-pumped, pulsed amplifier. The amplifier consists of a 114 mm long, 6.35 mm diameter Nd:YAG rod, a Xenon flashlamp and water cooled pump chamber. The double-pass gain is \(10^3\).

The resulting, approximately 150-200 microsecond duration pulse is formed into a “burst” train using a custom, dual Pockels cell “slicer” (Medox Inc.). The train can have a variable number of pulses, between 1 and 99, with inter-pulse spacing as short as 1 microsecond (1 MHz repetition rate). The pulse slicing is achieved by rapidly rotating the polarization of the pre-amplified pulse by 90\(^\circ\) using one of the two Pockels cells. The application of a suitably high voltage to the KDP crystal in each of the Pockels cells causes a \(\lambda/4\) retardation of the beam for each pass, resulting in a 90\(^\circ\) rotation in polarization when double passed. In addition, the extraordinary axes of the two KDP
crystals are aligned 90° relative to one another. Although subtle, this alignment is necessary to provide cancellation of the $\lambda/4$ retardation between the two cells during the transition from ‘on’ to ‘off’ and vice-versa. When both Pockels cells are ‘off’ or ‘on’, the overall rotation is 0° and the beam does not get passed through the remaining amplifier chain. Although possessing fast rise times (~3 nsec), the slow recovery time (order of 100 nsec) of a single Pockels cell necessitates the use of a second cell to produce a sharp cutoff; this allows formation of individual pulses as short as 6 nsec.

Figure 3.1: Schematic Diagram of Pulse Burst Laser.
The burst of pulses formed by the Pockels cells is further amplified via a pair of double-pass flash lamp pumped amplifiers, Amplifier 2 and Amplifier 3. The pulses are then further amplified by Amplifier 4, a dual-flashlamp amplifier with 114 mm long/9.52 mm diameter Nd:YAG rod. Following the 4th amplifier, the pulses are reflected back by a phase conjugate mirror (PCM), whose function will be described shortly. After the second pass of the 4th amplifier, the pulses are finally amplified by the Amplifier 5, a dual-flashlamp, single-pass amplifier with 114 mm long/12.7 mm diameter Nd:YAG rod. Due to the large gain of the overall system (order of 109), six Faraday optical isolators are used to isolate each amplification stage and to prevent/minimize amplified spontaneous emission (ASE) in the backwards direction. KTP or LBO non-linear crystals are used to generate the Second Harmonic wavelength of 532nm.

Figure 3.2: Typical burst train, pulses are separated by 5 µs and 7 mJ each at 532 nm.
Our laser system operates at a repetition rate of 5 Hz, e.g. there are 5 burst trains every second. Figure 3.2 shows a typical burst train at 532 nm taken from an oscilloscope. Note that the apparent tail of the pulses is from the high input impedance of the oscilloscope used to capture the burst train. The real pulse width (FWHM) is about 8 nsec. Pulses are separated by 5 microseconds and contain approximately 7 mJ each. The pulses seem more uniform in this relatively low energy than that of the higher energy cases, which will be described below.

3.1 Phase Conjugate Mirror

A significant improvement made in the 2nd generation design is the incorporation of a phase conjugate mirror (PCM). The PCM consists of a 25.4 mm diameter x 152.4 mm long optical cell, which is filled with a high index-of-refraction liquid, in this case a fluorocarbon known as FC-75 (3M Corp.). The PCM utilizes the principle of Stimulated Brillouin Scattering (SBS), which is a nonlinear optical phenomenon that produces a coherent beam at 180° (“backscattering”) if the pump beam input achieves a minimum intensity, termed the SBS threshold. As can be seen in Figure 3.1, the beam is focused into the PCM after the 4th amplifier with a 100 mm focal length lens.

When an intense laser beam of frequency ω₂ passes through a nonlinear material, a coherent acoustic wave at a frequency ωₙ and an optical beam at a frequency ω₂-ωₙ are produced simultaneously. For example, an optical wave E of frequency ω moving in (+z) direction is given by
\[ E(x,y,z,t) = A(x,y) \exp\{-i[wt + kz + \phi(x,y)]\} \quad (3.1) \]

After the PCM, a conjugated wave, Ec, propagates, given by:

\[ Ec(x,y,z,t) = A(x,y) \exp\{-i[wt - kz - \phi(x,y)]\} \quad (3.2) \]

The scattered optical beam has a reversed direction ("backscattering") and a reversed phase from the incident beam. This generation occurs only above a well-defined input threshold value. This phenomenon is called Stimulated Brillouin Scattering\(^{[42]}\).

The PCM serves two purposes. First, it eliminates the low intensity "pedestal" superimposed on the high intensity pulses which make up the desired output of the burst train. Second, it reduces ASE growth by providing additional isolation between the first three stages of amplification and the last two stages. PCMs have been used in other laser systems for the purposes of pulse narrowing\(^{[49, 50]}\) and suppression of ASE\(^{[51]}\), but have not previously been used to separate high intensity pulses from a low intensity background.

Even when nominally “off”, the Pockels cell switcher imparts a small phase retardation to the incident laser beam. The ratio of the ‘on’ signal to the ‘off’ signal is known as the contrast ratio and was measured to be approximately 2000:1. The residual 0.05% light is passed on through the remaining amplifiers in the system and forms a pedestal upon which the desired pulses sit. Since the Pockels cells are “on” for only \(~10\) nsec (duration of single pulse) and “off” for \(~10\) microseconds (nominal separation between pulses), this results in substantial integrated energy in the pedestal. In addition, it appears that the pedestal is preferentially amplified, relative to the pulses, in the later stages of amplification. The incorporation of the PCM, however, acts as an intensity
filter whereby the high intensity of the desired pulses exceeds the threshold SBS intensity and reflects from the PCM while the lower intensity pedestal remains below the threshold and passes through the PCM and is rejected. Thus, the energy contained in the final amplification stages is transferred exclusively to the pulses and not the pedestal.

The PCM also has the benefit of reducing ASE. By analogy to a voltage amplifier, ASE occurs when the gain is sufficiently high that significant laser system output occurs in the absence of any input from the master oscillator. Similar to the pedestal described above, the intensity of the ASE component before the PCM is well below threshold and, as a result, it is not reflected by the PCM into the system. The build-up process must then start over with the spontaneous emission from the fourth and fifth amplifiers.

The benefits of the PCM are best illustrated through Figure 3.3, which shows the average power output of the laser at 532 nm as a function of the average fundamental power. Conversion to 532 nm is achieved using a type II KTP crystal. The non-linear conversion to 532 nm is intensity-dependent whereby the conversion efficiency is poor at low intensities and quite efficient (up to 50%) for higher intensities. Thus, the fraction of light converted to 532 nm from 1064 nm is a good indication of the amount of energy contained within individual pulses. The first point on both curves in Figure 3.3 corresponds to operation of the laser with amplifiers #4 and #5 turned off. Each subsequent power measurement was made by incrementally increasing the energy supplied to the flashlamps by 10 J, beginning with amplifier #4 and proceeding to amplifier #5 after #4 reached its maximum output of 74 J. It should be noted that amplifier #5 was only operated at 55% of capacity for the measurements performed.
without the phase conjugate mirror in order to avoid potential damage to the KTP crystal. By this point, however, the resulting trend and benefit of the PCM is quite clear.

![Figure 3.3: Average second harmonic power as a function of average fundamentals with (diamonds) and without (squares) incorporation of Phase Conjugate Mirror.](image)

At the lowest power setting (the leftmost point on each curve), which are obtained with the same flashlamp energies, more 532 nm power is actually produced without the PCM (55 mW) than with the PCM (38 mW). This reveals substantial losses of pump power (1064 nm) imparted by the PCM when operated near the SBS threshold. Thus, for relatively low amplification following the PCM, the effect of the PCM is to decrease the overall system power. As the post-PCM amplification is increased, however, the amount of 532 nm energy increases at a substantially higher rate when the PCM is included. This
is particularly true at the highest power settings where the PCM produces average power at 532 nm almost 7X larger than without the PCM. The overall conversion efficiency from 1064 nm to 532 nm reaches a maximum of only ~6% without the PCM. Inclusion of the PCM increases the harmonic conversion efficiency greatly, reaching a maximum of ~50%. Figure 3.3 was produced for an arbitrary pulse burst setting of 8 pulses with inter-pulse timing of 10 microseconds (100 kHz repetition rate) although very similar results were achieved for other pulse settings. In general, the benefit of the PCM increases for bursts containing a greater number of pulses as the pulses must compete with each other for gain as well as with the pedestal.

3.2 Second Harmonic Generation

In the Pulse Burst laser system, the primary oscillator is a CW 1064 nm Nd:YAG laser. By using a 10×10×12 mm KTP Type II crystal or a 7×7×20 mm LBO Type I crystal, we can generate the second harmonic wavelength (532 nm). As discussed in Chapter 2, the theoretical SHG conversion efficiency\[42\] is given by:

\[
\frac{P^{(2\omega)}}{P^{(\omega)}} = 8\varepsilon_0^{1/2} \mu_0^{3/2} \frac{n^2 d_{\text{eff}}^2 L^2}{A} \left(\frac{P^{(\omega)}}{A}\right) \frac{\sin^2(\Delta k L / 2)}{(\Delta k L / 2)^2} \tag{3.3}
\]

where \( L \) is the length of the nonlinear crystal, \( A \) is the area of the fundamental beam, \( d_{\text{eff}} \) is the effective nonlinear coefficient, and \( \Delta k = k^{(2\omega)} - k^{(\omega)} \). The last term in
equation (3.3) is a sinc function: \( \text{sinc}(x) = \frac{\sin(x)}{x} \). It has the maximum value when \( x = 0 \), so we need to make \( \Delta k = 0 \), the Phase-Matching condition, in order to obtain the highest conversion efficiency.

The Phase-Matching condition for Second Harmonic generation is \( k^{(2\omega)} = 2k^{(\omega)} \).

If we use the relation \( k^{(\omega)} = \omega \sqrt{\mu e_0 n^{(\omega)}} \), it becomes \( n^{2\omega} = n^{(\omega)} \). As discussed in Chapter 2, the refractive index \( n \), which a light beam experiences in a non-linear crystal, depends on the polarization direction. For a uniaxial crystal, which has only one optical axis, there exists two cross sections of the index surface, a sphere for ordinary waves with index \( n_o \), and an ellipsoid for extraordinary waves with index \( n_e(\theta) \). The optical axis is the direction for which \( n_e = n_o \). For a negative \( (n_e < n_o) \) uniaxial crystal, if \( n_e^{2\omega} < n_o^{2\omega} \), there exists an angle \( \theta_m \) at which the Phase-matching condition is satisfied.

There are two types of processes in harmonic generation \[^{23}\] , depending on the two possible orientations for the linear polarization vectors of the incident beams. In the Type-I process both incident beam polarization vectors are parallel; in the Type-II process the polarization vectors are orthogonal.

\[
n^{2\omega}(\theta_m) = n_o^{2\omega}, \text{ Type-I}; \quad n^{2\omega}(\theta_m) = \frac{1}{2} [n_e^{2\omega}(\theta_m) + n_o^{2\omega}], \text{ Type-II} \tag{3.4}
\]
A. Critical Phase Matching

If phase matching is accomplished at an angle $\theta_m$ other than 90° with respect to the optical axis of a uniaxial crystal, there will be double refraction. Therefore, the direction of power flow of the fundamental and second harmonics will not be completely collinear but occur at a small Walk-off angle. For a negative uniaxial crystal and Type-I phase matching, this angle is given by:

$$\tan \rho = \frac{(n^o_{\omega})^2}{2 \left( \frac{1}{(n^e_{2\omega})^2} - \frac{1}{(n^o_{2\omega})^2} \right) \sin(2\theta_m)}$$

(3.5)

The angle $\rho$ has the effect of limiting the effective crystal length over which harmonic generation can take place. The beams completely separate at a distance of order

$$l = a / \rho$$

(3.6)

called the aperture length, where $a$ is the beam diameter. Of course, at only a fraction of this distance the reduction of conversion efficiency due to walk-off becomes noticeable and has to be taken into account.

B. Non-critical Phase Matching

If the refractive indices can be adjusted so that $\theta_m = 90^\circ$, there are no Walk-off effects due to double refraction. This is called Non-critical phase matching, which can be
achieved by temperature tuning of the crystal. We applied this Non-critical phase matching for our SHG and THG using the Type-I LBO crystals.

C. Non-linear crystals

Several non-linear crystals are discussed in this dissertation, for which a list of the most important parameters are given in Table 3.1 below. The parameter \(d_{\text{eff}}\) is the nonlinear coefficient, which relates the field amplitude generated by the non-linear process (for example, the second harmonic) to the field amplitude of the “pump”. Since \(I \propto E E^*\), non-linear conversion is typically proportional to \(d_{\text{eff}}^2\) (see equation 3.3).

<table>
<thead>
<tr>
<th></th>
<th>LBO</th>
<th>BBO</th>
<th>KTP</th>
</tr>
</thead>
<tbody>
<tr>
<td>(d_{\text{eff}} \text{ [pm/V]})</td>
<td>1.16</td>
<td>1.94</td>
<td>3.64</td>
</tr>
<tr>
<td>Transparency [(\mu\text{m})]</td>
<td>0.16-2.6</td>
<td>0.19-2.5</td>
<td>0.4-3.5</td>
</tr>
<tr>
<td>Damage Threshold ([\text{GW/cm}^2])</td>
<td>2.5</td>
<td>1.5</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Table 3.1: Parameters of common non-linear crystals.

However, for instantaneous high power pulsed laser beams, material damage threshold is often as important (or more important) as \(d_{\text{eff}}\). As can be seen in Table 3.1, among these three crystals, LBO has higher damage threshold, but lower non-linear coefficient. KTP is the opposite, exhibiting lower damage threshold and higher non-linear
coefficient. They are both good for use in Second Harmonic Generation at 532 nm, but we needed a longer LBO crystal than KTP to obtain the same conversion efficiency. Since the Third Harmonic wavelength of Nd:YAG laser is 355nm, KTP is not good because it is opaque at this short wavelength. BBO and LBO have good transparency in UV range. BBO is widely used in OPO cavities with 355nm pumping because it has a higher non-linear coefficient than LBO. BBO is also widely used as mixing crystal to generate UV light.

D. Burst sequences at 532 nm

Figure 3.4, 3.6, and 3.7 show some typical high power 532 nm burst sequences. Again, the apparent tail of the pulses is from the high input impedance of the oscilloscope used to capture the burst train.

Figure 3.4 shows a typical 532 nm burst sequence of 6 pulses with 10 µs spacing. The average pulse energy is 88 mJ.

Within a burst of pulses, the energy can vary considerably between pulses. It is important to point out that the ability to obtain pump burst envelopes with relatively uniform individual pulse energy is extremely important for successful burst mode operation of the second harmonic generation, third harmonic generation, and especially for OPO. The profile in Figure 3.4 shows that the energy varies across the burst with 1.5:1 ratio between the highest and lowest energy pulses. Upon non-linear conversion to the third harmonic and OPO output, this non-uniformity will increase. The distribution of energy is dependent upon the number of pulses, pulse separation, and amplifier delay.
Figure 3.4: Typical 0.532 micron sequence of 6 pulses with 10 usec spacing. Average pulse energy is 88 mJ.

In general, the pulse energy distribution resembles the 100 to 150 µs gain curve associated with each of the flashlamp-pumped amplifiers, as can be seen from figure 3.5. As more energy is added to each amplifier, this time duration will be shorter because gain narrowing occurs. Figure 3.5 shows the small signal gain of each amplifier. We measured the gain curve by double passing the CW laser directly into each amplifier. In many cases, as the amount of time between pulses is increased, the partially depleted gain will grow back during the interpulse period and higher individual pulse energies can be obtained. This is limited, however, by the 100 to 150 µs window in which the flashlamps impart significant energy to the laser rods.
In general the total amount of energy within a burst of pulses is approximately conserved, regardless of the number of pulses and/or interpulse timing. For example, at one power setting and a pulse burst setting of 4 pulses separated by 8 microseconds each, the total 1064 nm energy measured was 120 mJ/burst, or an average of 15 mJ/pulse. Changing the pulse burst setting to 8 pulses separated by 4 microseconds each, the total amount of energy remained at 120 mJ/burst, but the individual pulse energy lowered to 7.5 mJ/pulse. During typical experiments, we need to make the total time duration of the burst sequence shorter than the 100-150 µs amplification window. One approach is to set different delay times for the five amplifiers. However, the final output pulse energy will be lower with this way because the total gain is lower. An example is illustrated in figure 3.5.
3.5, in which the five amplifiers can be seen to have different temporal gain profiles (even though they are triggered by a single trigger signal). Amplifiers 4 and 5 have longer amplification FWHM than the other three amplifiers, due to differences in the current pulse which powers the flashlamp. It can also be seen that the peak gain of Amp 4 comes \( \sim 150 \, \mu s \) after Amp 5 and that Amps 1, 2, and 3 have similar gain curves. (In our experiments, we normally delay these three amplifiers more than the other two.) Since the uniformity of the burst envelope is extremely important, adjustment of the gain curves is essential for OPO generation. We use a photodiode and oscilloscope to detect the burst envelope. The individual amplifier delay times are varied until the most uniform burst sequence is obtained.

![Figure 3.6: Typical 0.532 micron sequence of 8 pulses with 8 usec spacing. Average pulse energy is 60 mJ.](image)
Figure 3.6 indicates a typical 532 nm burst sequence of 8 pulses with 8 µs spacing. The average pulse energy is 60 mJ. Figure 3.7 shows a typical 532 nm burst sequence of 10 pulses with 5 µs spacing. The average pulse energy is 54 mJ. In addition to these burst sequences, we have also demonstrated burst sequences of 1 – 30 pulses, with inter pulse separation in the range 4 – 12 microseconds, although the individual pulse energy is lower in some cases.

![Graph showing intensity vs time for a typical burst sequence](image)

Figure 3.7: Typical 0.532 micron burst sequence of 10 pulses with 5 usec spacing. Average pulse energy is 54 mJ.

### 3.3 Third Harmonic Generation

Figure 3.8 shows the optical setup to generate the third harmonic wavelength (355nm). We use the same approach described by Dergachev, et al. [52]. Two 7×7×20
Type I non-critical phase matched LBO crystals are used for SHG and THG. Both crystals are heated up to \(\sim 40^\circ C\). The 1064 nm laser beam inputs into the SHG crystal after a 1.5:1 telescope. The output 532nm beam is generated with polarization perpendicular to the 1064 nm fundamental beam, as described in the last section. This needs to be rotated back for the THG step, which is accomplished using a 532 nm half wave-plate. The second harmonic beam is separated from the fundamental beam by a 45° 532 nm mirror, and then is reflected into THG crystal by another 532 nm mirror after its polarization is converted. Although these two mirrors are not dichroic mirrors, they have good transmission at the fundamental wavelength 1064 nm, more than 95%. The fundamental and second harmonic beams, which have the same polarization, are then mixed in the THG crystal. The output is the third harmonic, 355nm beam.

Figure 3.8: Schematic diagram of Type I Third Harmonic Generation.
The SHG and THG conversion efficiencies using the pulse burst laser are summarized in Figure 3.9 which plots conversion efficiency versus the fundamental input pulse energy. The results in Figure 3.9 are based on the burst sequences with 6 pulses and 10 µs interpulse spacing. Peak efficiencies are above 50% for SHG and 40% for THG. The trend of both the SHG and THG curves indicates that even higher conversion efficiencies can be achieved as the efficiency continues to rise with increasing input energy.

Figure 3.9: Second (squares) and third (diamonds) harmonic conversion efficiency as a function of average fundamental pulse energy.
Figure 3.10 indicates a typical fundamental, second harmonic, and third harmonic pulse sequence, which corresponds to the maximum energy data in Figure 3.9. The 6-pulse burst sequence has an average individual pulse energy of 130 mJ at the fundamental wavelength 1064 nm, 66 mJ at 532 nm and 52 mJ at 355 nm. The burst envelopes are reasonably uniform.
Figure 3.10: Typical Fundamental (a), second harmonic (b), and third harmonic (c) pulse sequences. Average individual pulse energy at 355nm is 52 mJ. Conversion efficiency is $\sim 40\%$. 
Figure 3.11 shows a typical 8-pulse, 5 µs interpulse spacing burst sequence at 355 nm. The average pulse energy is 35 mJ. The burst envelope is more uniform than that in figure 3.9. The energy ratio between the highest and the lowest pulses is only ~1.2:1.

![Figure 3.11: Third harmonic burst sequence with 8 pulses, separated by 5 µs. Average pulse energy is 35 mJ.](image1)

Figure 3.12: Third harmonic burst sequence with 10 pulses, separated by 4 µs. Average pulse energy is 26 mJ.

![Figure 3.12: Third harmonic burst sequence with 10 pulses, separated by 4 µs. Average pulse energy is 26 mJ.](image2)
Figure 3.12 indicates a typical 10-pulse, 4 µs interpulse spacing burst sequence at 355nm. The average pulse energy is 26 mJ. The burst envelope is also very uniform. In figure 3.10 — 3.12, the total time duration of the burst sequences are approximately the same, 36 µs. The individual pulse energy is less when there are more pulses in a burst train, but the total energy of each burst is still about the same, ~ 280 mJ/burst.

To summarize, we have demonstrated the capability of producing second harmonic and third harmonic, 6-10 pulse/burst sequence with good conversion efficiencies, reasonably uniform burst envelopes and high single pulse energies (30-50 mJ). Such burst sequences will now be used for PDV experiments and will be used to pump the OPO system.

3.4 Development of and Application of MHz PDV

An important application of the pulse burst laser is the newly developed experimental diagnostic technique, termed MHz rate planar doppler velocimetry (PDV). PDV has been developed over the last decade and a half and is now an accurate and viable technique used in many laboratories for planar velocity measurements in high-speed flows. Prior to this work at OSU, all PDV measurements had employed either CW lasers, which give time averaged data, or commercial single pulse “Q-switched” lasers, which give instantaneous data but at 10 Hz repetition rate. High repetition rate PDV has not been explored due to limitations of available lasers. With the development of the pulse burst laser however the capability now exists to apply PDV at MHz rates. Section 3.4 describes works performed collaboratively with Brian Thurow and Marc Blohm, two
former students of the GDTL, which has resulted in the first experimental diagnostics capable of obtaining quantitative planar velocity data at MHz frame rates.

3.4.1 Introduction

Planar Doppler Velocimetry (PDV) is a powerful optical diagnostic technique that can be used to measure all three components of instantaneous velocity over a two-dimensional plane within a flow field with high spatial resolution. PDV accomplishes this task by measuring the Doppler shift in frequency of light scattered by moving particles in the flow field. The relatively small frequency shift is discriminated using an atomic or molecular vapor filter. The Doppler shift, $\Delta f_d$, is related to the fluid velocity by the simple expression,

$$\Delta f_d = \frac{(\vec{s} - \vec{o})}{\lambda} \cdot \vec{V}$$  \hspace{1cm} (3.7)

where $\vec{s}$ is the unit vector in the observation direction, $\vec{o}$ is the unit vector in the direction of the incident laser light, $\lambda$ is the wavelength of the light and $\vec{V}$ is the velocity vector of the flow.

The concept of measuring fluid velocity by means of the Doppler shift was originally utilized by Cummins et al.\textsuperscript{53} and Yeh and Cummins\textsuperscript{54} in 1964, in which the point measurement technique, now known as Laser Doppler Velocimetry (LDV)\textsuperscript{55}, is based. Subsequently, Komine and Brosnan\textsuperscript{56} and Meyers and Komine\textsuperscript{57} developed a
planar velocimetry method referred to as Doppler global velocimetry (DGV) where a molecular filter was used to measure the Doppler shift. This approach is conceptually similar to what is now known as Filtered Rayleigh Scattering [Miles and Lempert]. Since these original works, the technique has been further developed by numerous research groups including Elliott et al., McKenzie, Smith et al., Clancy et al., Arnette et al., Beutner et al., Mosedale et al. and Crafton et al.. In the course of development, many researchers began using the term planar Doppler velocimetry (PDV) to describe the technique.

Today, a typical one-component PDV instrument utilizes a pulsed injection-seeded Nd:YAG laser, two CCD cameras and a molecular iodine filter. The laser is used to illuminate a plane of the flow with narrow spectral linewidth light. The Doppler shifted scattered light is then split into two paths using a beamsplitter and imaged onto the cameras. In this manner the absolute absorption of scattered light, as it passes through an iodine cell placed in one of the beam paths, is measured at every spatial location within the object plane. For scattering by relatively large (as compared to molecular dimension) particles, this absorption is a function of particle velocity only. Accurate calibration and image mapping algorithms have been developed with the result that velocity accuracies of ~1-2 m/s are now achievable. More details concerning the history of PDV, the art of its application and recent advances can be found in comprehensive review articles by Samimy and Wernet and Elliott and Beutner.

A current limitation, common to any planar velocity measurement technique, is its inability to acquire time-correlated sets of velocity data in high-speed flows. As discussed earlier, typical commercially available high energy pulsed Nd:YAG lasers are
limited to repetition rates of the order of 10 Hz and only recently have ultra-high frame rate digital cameras become commercially available. These factors have limited PDV to instantaneous and ensemble-averaged flow measurements. The development of the MHz rate pulse burst laser system allows for the extension of the PDV technique to high-repetition rates.

3.4.2 Experiment

Figure 3.13 shows a schematic of the experimental arrangement used for obtaining one-component PDV data.

![Schematic of one-component PDV experimental arrangement.](image)

Figure 3.13: Schematic of one-component PDV experimental arrangement.
Scattered laser light from the flow field initially passes through a polarizer. The polarizer serves two purposes. First, it helps minimize background scattering of light, which tends to be depolarized. Second, it removes any dependence of the beamsplitter on incident light polarization by ensuring that all light entering the system is of the same polarization. The scattered light is then split into two paths, a signal path and a reference path, using a 50/50 beamsplitter. The signal path goes directly through the beamsplitter and subsequently through the iodine molecular filter after which it is imaged onto a CCD camera, termed signal camera. The reference image is reflected by the beamsplitter and an additional mirror. After passing through a neutral density (ND) filter, it is imaged onto a 2nd CCD camera, termed the reference camera. The ND filter is used to balance the intensity of the signal and reference images. During experimental runs, the arrangement was enclosed in a box to prevent flow of air through the system and to block any ambient light.

Figure 3.14: Schematic of frequency monitoring system.
Since the flow velocity is based on the frequency shift, accurate measurement of frequency shift is extremely important. As discussed earlier, the master oscillator of our pulse burst laser is a CW narrow linewidth Nd:YAG laser. The center frequency is determined by the temperature-dependent index-of-refraction and geometry of the Nd:YAG crystal within the cw laser and is tuned by adjusting the voltage input to a thermoelectric cooler in contact with the crystal. Across the entire voltage tuning range of -10 to +10 V, the tuning coefficient was measured to be 8.62 GHz/Volt. The pulse burst laser does not experience the pulse-to-pulse frequency shift jitter that plaques commercial Nd:YAG lasers, but the laser frequency can drift due to thermal expansion/contraction of the ring cavity. Although a fan is used for cooling, small variations of temperature still occur, leading to a slow frequency drift which was measured to be ~50 MHz/hour.

A frequency monitoring system is used during the experiment to monitor this laser frequency drift, as shown in figure 3.14. A low energy (~0.5%) portion of the beam is sampled from the main beam using a thin film polarizer and a half wave plate. The beam is expanded to ~20 mm diameter where it enters the frequency monitoring system. One half of the beam is passed through an iodine filter, focused onto a flash-opal diffuser and sampled by a high-speed photo-detector. The other half of the beam is likewise focused onto a diffuser and sampled by a photo-detector. Stanford Research Systems gated integrators are used to sample the photo-diodes output over an approximately 50 ns window timed to coincide with the first laser pulse within a burst of pulses. The resulting signal (filtered) and reference (unfiltered) measurements are then compared to determine the amount of absorption produced by the iodine cell.
Figure 3.15 shows filter profiles obtained by scanning the tuning voltage of the pulse burst laser from 7.9 to 8.6 volts in 0.01 V increments and averaging 100 measurements for each point. In our experiments, the laser frequency was set to yield a transmission ratio of ~0.2 (~8.35 V in Figure 3.15), which corresponds to a ratio of 0.23 in the frequency monitoring system.

![Figure 3.15: Transmission ratio of iodine molecular filters vs. laser tuning voltage.](image)

During the experiment, two CCD cameras capture the flow images simultaneously, one with filter and one without filter. The two images see exactly the same region of the flow, but the intensities differ, depending upon flow velocity. We can compare every image set and obtain the transmission ratio of every pixel of these flow images. After the ratios are established, the velocity can be easily determined.
Consequently, lower velocities yield a lower transmission ratio and high velocities a higher transmission ratio. A look-up table incorporating interpolation and the profile shown in Figure 3.15 is used to determine the corresponding frequency shift (given in volts and converted to frequency using 8.62 GHz/V) at each pixel location. Using this measured frequency shift, the velocity can be determined from equation 3.7.

3.4.3 Results

The CCD cameras used in these experiments are PSI-IV cameras from Princeton Scientific Inc. The cameras, which will be described in more detail in Chapter 5, can acquire 28 images at a variable rate as fast as 1 MHz.

![Figure 3.16: A typical burst sequence used for PDV experiment with 28 pulses and 4 µs spacing.](image)
Figure 3.16 shows a typical burst train used for a PDV experiment, which contains 28 pulses with 4 µs interpulse spacing. The individual pulse energy is ~ 2mJ. Since Mie scattering serves as the basis for the detected signal in a PDV experiment, this pulse energy is sufficient for the cameras to obtain clear flow images. In fact, during our experiments this energy was sufficient to saturate our cameras in some cases in which acetone was seeded into the core flow. The burst envelope has an energy ratio about 2.5:1 between the highest and the lowest pulses. In fact, this ratio is pretty good considering the use of as many as 28 pulses. We can achieve this uniformity because the individual pulse energy is relatively low, so we can better adjust the relative delay times of the individual amplifiers.

Figure 3.17 gives a typical example of a PDV velocity image sequence, which displays 28-frame obtained from a Mach 2 nozzle. The diameter of the nozzle exit is 1". The ambient pressure is 1 atmosphere. The flow is air and acetone is seeded in the core flow. Images are arranged left-to-right, top-to-bottom and 4 microseconds separate each frame. The entire set of 28 frames is included in figure 3.17 to emphasize the ability of the MHz rate PDV system to acquire data at high frame rates. Observing the images from top-to-bottom may aid the reader in following the motion of turbulence structures across the image sequence. As an example, a circle in the 2nd frame of the sequence highlights a patch of low-speed (darker shading) fluid that is pulled and stretched into the high-speed jet core. Along the centerline of the jet in Figure 3.17, the measured velocity component is in the neighborhood of 340 m/s, which would correspond to a centerline
speed of ~500 m/s with this experimental setup. For an ideally expanded Mach 2.0 jet, the centerline velocity would be ~505 m/s.
Figure 3.17: A sequence of 28 velocity images.
In this chapter we will demonstrate the ability to generate trains of tunable wavelength pulses using a custom built Optical Parametric Oscillator (OPO), which is the first step toward our goal of developing an ultra-high frame rate Planar Laser Induced Fluorescence (PLIF) imaging system. We will compare and contrast second and third harmonic pumping of the OPO. We will also describe some analysis of the narrowing of the OPO spectral linewidth, by using injection seeding.

4.1 Third Harmonic Pumping

The third harmonic pumped OPO cavity, which is shown in Fig. 4.1, is home built and was designed specifically for use with the burst mode Nd:YAG pump laser. The OPO “gain” medium consists of a pair of 12 mm (length) x 5 x 7 mm (cross section) Type I BBO crystals, arranged in a linear cavity configuration. The crystals are oriented to provide walk-off compensation between the pump and signal/idler beams, which provides higher effective gain for the relatively small cross sectional area (~ 2-4 mm$^2$) pump beam. We tried two configurations in our experiments, a singly resonant OPO
cavity, and a doubly resonant OPO cavity. For SRO case, only the signal wave at 622 nm is resonant in the cavity. In other words, the high reflector mirror in figure 4.1 only has high reflectance at the signal wavelength. For the DRO case, the cavity is doubly resonant on the signal and idler wavelengths, using a broad-band high reflector and output coupler coated for 20 – 30% reflectivity in the wavelength range 600 - 850 nm. The 355 nm pump beam is coupled into and out of the cavity using a pair of 45° dichroic mirrors, which reflect the 355 nm pump beam and transmit the OPO signal and Idler beams. By using a 355 nm 0° reflection mirror, the 355 nm pump beam can be retro-reflected back through the OPO cavity for a second pass. Note that double pass alignment requires very low angular displacement due to the phasing-matching requirement. More specifically, angular displacement of the retro-reflected pump is equivalent to changing the BBO crystal angle. Since a 1° angle change affects the OPO signal wavelength by ~70 nm, it is clear that careful alignment is required.

Fig. 4.1: Schematic diagram of OPO Cavity illustrating injection-seeding and double pass of 355 nm pump beam. Cavity is doubly resonant at signal and idler wavelengths.
The total cavity length is \( \sim 10 \) cm, limited, currently, by the somewhat large pump mirror mounts. A single frequency external cavity diode laser is used to injection seed the cavity at either the signal or idler \(^{70}\) wavelengths, depending upon the experiment.

The seed radiation is injected through the output coupler using a Faraday rotation optical isolator, in a manner similar to that employed previously for injection seeding of a Titanium:Sapphire laser \(^{71}\). Two more low power optical isolators, not shown in Fig. 4.1, are used to prevent feedback-induced damage to the diode laser. Signal and idler wavelengths are separated using a dichroic mirror, also not shown in Fig. 4.1.

4.1.1 OPO Conversion Efficiency

Our pump beam pulsewidth is \( \sim 10 \) ns, which corresponds to \( \sim 10 \) feet in length, since the speed of light is \( \sim 1 \) foot/nsec. To increase OPO output efficiency, therefore, we need to make the optical cavity as short as possible. The single-pass power gain \( G \) of the parametric amplifier produced by a pump beam of intensity \( I_p \) can be approximated by \(^{23}\):

\[
G = \frac{1}{4} \exp\left(2l \frac{8\pi^2 d_{\text{eff}}^2 I_p}{\lambda_s \lambda_s n_s n_p \varepsilon_0 c} \right)
\]

(4.1)

where \( l \) is the crystal length, and \( d_{\text{eff}} \) is the effective nonlinear coefficient. Here, we define \( \varepsilon_0 d \) instead of \( d \), as in chapter 2, as the nonlinear coefficient. In this case, \( d \) has units of \( m/V \). For BBO, \( d \) is equal to 1.98pm/V. For example, if the pump 10ns pulse is 30 mJ with a beam diameter of 3.6mm, the pump intensity \( I_p \) is \( 2.4 \times 10^7 \) W/cm\(^2\). If we use a 12mm long BBO crystal, the single-pass gain is 3.4; but for two crystals with walk-off
compensation, the single pass gain increases to 11.5. These numbers are 46 and 2116, respectively, if we use a 1.8 mm diameter beam with the same energy.

The theoretical threshold intensity of a doubly resonant OPO system (DRO) is given by:

\[
I_{th} = \left( \frac{1}{\varepsilon_0 \mu_0} \right)^{1/2} \frac{n_p n_s n_i (g_l)^2}{8 \omega_s \omega_r i^2 d_{eff}^2}
\]  

(4.2)

where \((g_l)\) satisfies the threshold condition: \(\cosh(g_l) = \frac{1 + R_s R_l}{R_s + R_i}\), \(R_s\) and \(R_i\) is the reflectivity of the output coupler for the signal wave and idler wave. For a singly resonant OPO (SRO), \(R_i\) is equal to zero. For example, if we use two 12 mm long BBO crystals in a SRO system with a 20% reflectivity output coupler, the threshold pump intensity is \(\sim4.5\text{MW/cm}^2\). For a 10ns pulse with a beam diameter of 3.6mm, this threshold corresponds to 5.7 mJ.

![Figure 4.2: OPO conversion ratio vs. cavity length.](image-url)

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Another factor that can affect the conversion efficiency is the OPO cavity length. A shorter cavity results in more passes within the cavity during the short (~10nsec) pump pulse, which will result in higher conversion efficiency. Figure 4.2 shows the OPO conversion efficiency as a function of cavity length. The data are based on a SRO cavity pumped by a Continuum Surelite 10 Hz Nd:YAG laser.

A. Conversion Efficiency of Singly Resonant OPO

First, to study the characteristics of OPO, we built a singly resonant OPO (SRO) system. It is a “signal” wave resonant cavity. The high reflector has ~100% reflectance at the signal wavelength, 622 nm, and the output coupler reflects 20% at this wavelength.

![Figure 4.3. Conversion Efficiency from 355 nm Pump to Signal at 622 nm](image_url)
Figure 4.3 shows the single pulse conversion efficiency data obtained using the burst mode laser with 355nm pumping. We use two and three 7\times5\times12 mm BBO type I crystals to measure the conversion ratio. The pump beam diameter is about 3.5mm and OPO output is 622 nm. The cavity lengths for 2 and 3 crystals are 114 and 127 mm, respectively. It can be seen that operation with three crystals results in a lower threshold than operation with two crystals, but in both cases, the conversion efficiencies converge to about the same value \(\sim 28\%\), at high power (\(\sim 35\)mJ/Pulse). We also find that the conversion ratio rises rapidly with increasing pump energy.

The three-crystal OPO configuration output higher conversion efficiency at low energy pumping than the two-crystal configuration. However, there are some disadvantages for three-crystal OPO operation. First, the saturation conversion efficiency is about same as for two-crystal OPO, which can be seen from figure 4.3. Second, you cannot obtain walk-off compensation from a three-crystal OPO, as there is no compensation for the third crystal. At this point, a fourth crystal could be put in the OPO cavity to make the compensation, but the conversion efficiency will be balanced by the longer cavity length. Therefore the OPO cavity with more than two crystals doesn’t result in higher realized conversion efficiency. Based on these results we decided in subsequent experiments to use two crystals, in a doubly resonant OPO configuration.

B. Conversion Efficiency of a Doubly Resonant OPO

Based on the experience from the operation of the singly resonant OPO, we built the doubly resonant OPO (DRO) system, shown in figure 4.1. Here, the term DRO cavity means that both signal and idler waves can resonant in the cavity. It does not mean that
the cavity length has been carefully stabilized to match a single longitudinal mode for both signal and idler waves simultaneously. This will be discussed in more detail in section 4.3.

Figure 4.4 shows combined OPO signal + idler power conversion efficiency as a function of input pulse energy when the OPO cavity is operated in single and double pass pump configurations. For these measurements the burst mode laser was used in a manner identical to that used to generate high frequency pulse trains, but with only a single pulse. It can be seen that operating in double pass pump mode (see figure 4.1) reduces the OPO threshold by approximately 50%, from ~8 to ~4 mJ. More importantly, it can be seen that the efficiency rises more rapidly as a function of pulse energy, reaching a plateau of ~36% for pump input energy of ~30 mJ. The plateau begins from 30% efficiency for pump energy of ~17 mJ. This is in contrast to the single pass geometry in which the conversion efficiency rises rather slowly with increasing pulse energy, reaching a maximum of ~30% at the highest pump energy (~54 mJ) employed.

For comparison, the theoretical threshold for single pass pumping of a doubly resonant OPO cavity can be estimated from (4.2). Equation (4.2) predicts a threshold intensity of ~2.2 MW/cm², corresponding to a threshold pump pulse energy of ~0.75 mJ (for double pass) for our beam of nominal diameter 3.0 mm and pulse duration of 10 nsec, in quite good agreement with the observed threshold of ~4 mJ. (The difference is likely due to a combination of imperfect walk-off compensation, reflection losses from the non-coated BBO crystals and 355 turning mirrors, and the uncertainty in pump beam diameter.) Nonetheless we emphasize that achieving such low threshold implies that the beam spatial quality of the burst mode pump laser is quite good, an issue which was a
significant concern prior to performing these measurements. We also point out that low threshold, while always important for nonlinear processes, is particularly critical for burst mode operation of the OPO due to the inherent intensity envelope of the individual pump pulses within the burst. While we have not yet explored this in detail we note a general guideline for OPO operation which states that the pump intensity should be a factor of approximately four greater than threshold\textsuperscript{[23]}. As illustrated in figure 4.4, the plateau begins from \textasciitilde30\% efficiency for pump energy of \textasciitilde17mJ, which is about four times threshold.

![Figure 4.4: Conversion efficiency of OPO cavity shown in Fig. 4.1 for single 355 nm pulse from burst mode laser system.](image)
Because it is doubly resonant, the OPO output includes both signal (622 nm) and idler (827 nm) components with a measured signal idler energy ratio of 0.55:0.45, which is approximately equal to the ratio of single photon energies. In broad band operation the OPO was operated over a wide range of wavelengths from ~600/869 nm to 700/720 nm for the signal/idler respectively. It was found that the OPO output was approximately constant, at least over a “signal” wavelength range of ±50 nm. We also seeded the OPO system at idler wavelengths of 827 nm and 780 nm. In both seeding cases, the OPO conversion efficiency was not significantly changed, but the spectral linewidth was greatly narrowed.

4.1.2 Burst Sequences

In this section, we will show some burst sequences of the OPO output and input. We successfully demonstrated burst mode pumping for both singly resonant OPO and doubly resonant OPO. Since the conversion efficiency of the DRO system, shown in the last section, is better than that of SRO system, we focused on burst sequences using the DRO system.

A. Burst Sequences for Single Pass OPO Geometry

Figures 4.5-4.7 show some representative pump and OPO output burst sequences for single pass OPO pumping, obtained using an optical configuration identical to that used to obtain the single pulse-single pass data (the circles) in figure 4.4. Note that due to limitations in our sampling oscilloscope, the RC time constant has been increased (to a
few microseconds) in order to display the captured burst sequences. The relatively long observed decay of the individual pulses is an artifact of this and does not correspond to the actual (~ 10 nsec) individual pulse durations, as discussed earlier. It should also be pointed out that all of the following 355 nm pumping burst sequences and corresponding OPO output burst sequences are taken at the same time, so they record the development of the same burst train.

Figure 4.5 illustrates a burst sequence of six pulses, separated in time by ten microseconds. A variety of additional combinations have been produced ranging from 1 to 12 pulses with 4-12 µs spacing. Note that a significant effort was made to minimize the non-uniformity of the pulse energy distribution within the burst.

The average instantaneous pump energy for the burst sequence shown in figure 4.5 is ~34 mJ and the total conversion ratio is ~25%. In this case the OPO 622 nm signal energy is ~4.8 mJ per pulse.
Figure 4.5: Typical 6 pulse, 10 us spacing pump (upper) and single pass OPO output (lower) burst sequence. Total OPO conversion efficiency (signal + idler) is 25%. Average single pulse 355 nm pump energy is 34 mJ.
At the same conditions (34 mJ pump energy of figure 4.5), we find from figure 4.4 that the OPO conversion ratio is only ~21% for single pulse mode, which is somewhat lower than the observed burst mode case. This unexpected phenomenon was also observed when 8 and 10 pulse burst sequences were used. While the reason for this is not completely understood we speculate that the effect may be due to thermal lensing of the Nd:YAG rods. When we operate in high energy burst mode, increased thermal loading of the Nd:YAG rods cause the 355nm pump beam size to be somewhat decreased (~2.6mm for the 6 pulses case) compared to single pulse mode (~3.0 mm). It appears that this intensity difference may be sufficient to result in higher OPO conversion efficiency. Since the total energy supplied to the flashlamps is increased when the number of pulses in a burst sequence is increased, this makes the thermal lensing effect even stronger, as illustrated in the figure 4.6 and 4.7.

Figure 4.6 is very similar to figure 4.5, except that the number of pulses has been increased to eight. In order to keep the time duration about the same as that of the 6 pulse case, the time spacing between each pulse is decreased, to 8 µs. Once again, the input 355 nm pump burst is reasonably uniform. It is also noted that the intensity envelopes of the OPO output follow reasonably, but not precisely, the envelopes of the pump pulses. The energy ratios between the highest and the lowest pulses in pumping and OPO output burst sequences are about same, 1.5:1. The average pump energy is 24 mJ and the total conversion ratio is 26%, which is again higher than the ~15% observed for the single pulse case with this pulse energy. The OPO 622 nm signal energy is ~4.8 mJ per pulse in this case.
Figure 4.6: Typical 8 pulse, 8 us spacing pump (upper) and single pass OPO (lower) burst sequence. Total OPO conversion efficiency (signal + idler) is 26%. Average single pulse 355 nm input energy is 24 mJ.

Figure 4.7 illustrates another typical case, consisting of 10 pulses with 6 microsecond spacing. Again, both the pump and OPO output bursts are reasonably uniform and the intensity envelopes are very similar, which means each pulse has approximately equal conversion ratio. Although the average pump energy is only 18.6 mJ per pulse, the OPO conversion efficiency is 23%, which is significantly higher than that
observed for the single pulse case. The OPO 622 nm signal energy is ~2.4 mJ per pulse in this case.

Figure 4.7: Typical 10 pulse, 6 us spacing pump (upper) and single pass OPO (lower) burst sequence. Total OPO conversion efficiency (signal + idler) is 23%. Average single pulse 355 nm input energy is 18.6 mJ.
We note that ascribing the increase in conversion efficiency observed in burst mode operation to thermal lensing effects is preliminary and somewhat speculative. Other variables, such as alignment and beam spatial mode need to be more fully explored.

B. Burst Sequences for Double Pass OPO Geometry

Figures 4.8-4.10 show some representative double pass pump OPO pump and output burst sequences, obtained using an optical configuration identical to that used to obtain the single and double pass data (the squares) in figure 4.4. We again find that the OPO output intensity envelope reproduces well the respective pump envelope, indicating operation in the plateau region of figure 4.4, well above threshold.

Figure 4.8 shows burst sequences of 6 pulses with 10 microsecond spacing interpulse. The average pump energy is 31mJ/pulse and the average conversion ratio is 35%. In this case, the OPO output 622 nm energy is ~6.2 mJ per pulse. Figure 4.9 illustrates burst sequences of 8 pulses with 8 microsecond spacing between pulses. The average pump energy is 26mJ/pulse and the average conversion ratio is 33%. Similarly, Figure 4.10 shows burst sequences of 10 pulses with 6 microsecond spacing between pulses. The average pump energy is only 16mJ/pulse, but the average conversion ratio reaches 32%, indicating again an increase in conversion efficiency, albeit less than in the single pass pump case, compared to single pulse operation.
Figure 4.8: Typical 6 pulse, 10 us spacing pump (upper) and double pass OPO (lower) burst sequence. Total OPO conversion efficiency (signal + idler) is 35%. Average single pulse 355 nm input energy is 31 mJ.
Figure 4.9: Typical 8 pulse, 8 us spacing pump (upper) and double pass OPO (lower) burst sequence. Total OPO conversion efficiency (signal + idler) is 33%. Average single pulse 355 nm input energy is 26 mJ.
Figure 4.10: Typical 10 pulse, 6 us spacing pump (upper) and double pass OPO (lower) burst sequence. Total OPO conversion efficiency (signal + idler) is 32%. Average single pulse 355 nm input energy is 16 mJ.
A variety of additional combinations have been produced ranging from 1 to 12 pulses with 4-12 µs spacing. In all cases total signal + idler conversion efficiency is ~30% or more, essentially identical to that obtained using a single pulse. It is also noted that the intensity envelopes of the OPO output follow reasonably, but not precisely, the envelopes of the pump pulses, a result which we attribute to pump pulses with intensity significantly above threshold, by a factor of four or more. In this regard it is important to note that the pump laser produces bursts with an inherent intensity envelope, particularly when operated at high intensity where gain narrowing is significant. The multiple non-linear steps (2nd /3rd harmonic followed by OPO) will tend to exacerbate this problem and so it is critical to produce the most uniform pump bursts as possible.

4.2 Second Harmonic Pumping

To develop a better understanding of how to operate the OPO and how to potentially further increase the conversion efficiency, we also have studied a 532nm pumped system. We used a single crystal OPO cavity which was fabricated by Innovative Scientific Solutions, Inc., our industrial partner in this research program. The optical layout of the OPO cavity is shown in figure 4.11, which is a singly resonant cavity with the “signal” wavelength at 780nm. The 780 nm high reflection mirror is coated for high transmission at 532 nm, and the 20% output coupler is also coated for 532 nm. Only one 12 mm (length) x 5 x 7 mm (cross section) Type I BBO crystal is arranged in the cavity. With this setup the cavity length is minimized (51mm). We can also injection seed this
OPO system by using what is known as an external cavity diode laser (ECDL). The seeding setup is the same as that used for 355 nm pumping, and is shown in detail in figure 4.11. The seeding effect of the OPO system will be discussed in next section. The biggest difference between 532 nm pumping and 355 nm pumping is that at 532 nm it is possible to transmit the pump beam through the cavity end mirrors. This is not possible for 355 nm pumping due to coating damage by absorption of the UV beam. This geometry has the advantage that the cavity can be very short.

Figure 4.11: Schematic diagram of a single crystal OPO pumped by 532 nm.
We pumped this OPO with the second harmonic output of our pulse burst laser. The pump beam diameter is ~3 mm. Figure 4.12 shows the conversion efficiency based on single pulse pumping. Because the cavity is short, it can be seen that the conversion efficiency can be as high as 30% with only one crystal. We didn’t try pulse energies higher than 45 mJ because of concerns about mirror damage.

![Graph showing conversion efficiency vs. 532 nm pump energy](image)

**Figure 4.12:** Conversion ratio of 532nm pumped single BBO crystal OPO.

A few, albeit somewhat preliminary, burst sequences have been obtained using 532 nm pumping in the single crystal – minimum cavity length configuration described above. Figure 4.13 shows one example burst sequence consisting of 6 pulses with ten microsecond spacing. The average individual pulse energy is 32 mJ and average conversion efficiency to 780 nm signal is ~5%. While it is not yet clear why the average conversion efficiency is lower than that observed for the single pulse data (figure 4.12), it
is likely due to some combination of beam spatial mode, cross-sectional area, and divergence. It should also be pointed out that, unlike the 355 nm pumping cases illustrated in figs. 4.5-4.10, the burst to burst reproducibility for 532 nm pumping was somewhat unstable, often resulting in fewer than six readily observable pulses. This is an indication that the OPO burst illustrated in figure 4.13 corresponds to pump intensity in the vicinity of OPO threshold. Note also that as observed for 355 nm pumping, it appears that the conversion efficiency is somewhat higher for the first one or two pulses within the burst sequence. This is a further indication that the gain is lower for the subsequent pulses within the burst, possibly due to some sort of thermal effect. Here it is stressed that the results are preliminary. Our goal is to apply OPO to NO fluorescence, so we didn’t work hard to optimize 532 nm pumping.
Figure 4.13: Six pulses, 10 microsecond spacing burst sequences at 532 nm (upper) and 780 nm (lower). Average individual pump/signal pulse energies are 32 / 1.55 mJ, respectively.
4.3 Injection Seeding

Injecting a weak signal into a more powerful free-running oscillator can produce an interesting and useful set of injection locking effects. In 1865 Christiaan Huygens, while confined to bed by illness, noticed that the pendulums of two clocks in his room were invariably locked into synchronism if the clocks were hung close to each other, but became free-running when hung farther apart. He eventually traced the coupling mechanism to mechanical vibrations transmitted through the wall, thus providing one of the first observations of the coupling of two oscillators by injection locking. As described in reference \[72\], injecting a weak laser beam with narrow linewidth into a high intensity laser beam will “lock” it to the “seeding” frequency. This effect narrows the linewidth of the high intensity laser beam. For continuous wave (CW) injection locking of a CW OPO system, the full locking range in hertz frequency units is given by:

\[
\Delta v = -\ln(R_i) \frac{I_i}{\pi T} \sqrt{\frac{I_i}{I_o}}
\]  

(4.3)

where \( R_i \) is the reflectivity of the output coupler at the seeding wavelength, \( T \) is the round-trip transit time in OPO cavity, \( I_i \) is the injected signal intensity and \( I_o \) is the OPO output intensity of the signal or idler beam, which depends on signal seeding or idler seeding. For a pulsed OPO system, \( \Delta v \) is known to be much larger \[72\].

The experimental setup for OPO injection seeding is shown in figure 4.1, and a more detailed schematic diagram is given in figure 4.11. Although we use the same seed laser for the second harmonic pumped OPO and the third harmonic pumped OPO, the
injection seeding approach is different. For second harmonic pumping, we seeded the system at the “signal” wavelength, 780 nm, but for third harmonic pumping, we seeded it at the “idler” wavelength, as will be described in the next section.

4.3.1. Injection Seeding of a 355 nm Pumped OPO

As discussed earlier, a prime motivation for the work presented here is to develop the capability to perform NO-based Planar Laser Induced Fluorescence (PLIF) imaging at ultra-high frame rates. As such an ultimate goal is to mix the OPO signal output with residual 355 nm pump in order to generate tunable burst mode output in the vicinity of 226 nm. This will require OPO signal at 622 nm, a spectral region where it is difficult to obtain diode laser seed sources. A potential solution is to seed at the idler wavelength\[^{70}\], which for this case is \( \sim 827 \) nm.

A. Low Resolution Spectra

Figure 4.14 shows typical low-resolution spectra resulting from seeding the cavity at the idler wavelength of using an \( \sim 10\) mW 780 nm, ECDL (which is available in our laboratory). The spectra were obtained using an available 0.3 m spectrometer. Notice that both the signal and idler beams are narrowed significantly, from \( \sim 1 \) nm to a line width which is very difficult to measure using the low-resolution spectrometer.
Figure 4.14: Low resolution spectra of seeded and unseeded OPO signal 651 nm (upper) and idler 780 nm (lower) output. Note that the OPO is seeded at the idler wavelength.
Figure 4.15: Low resolution spectra of seeded and unseeded OPO signal 622 nm (upper) and idler 827 nm (lower) output. Note that the OPO is seeded at the idler wavelength.
During these experiments, it was found that idler seeding does not work for the singly resonant OPO cavity, whereas a doubly resonant cavity narrowed both signal and idler beams significantly. It was also determined that injection seeding resulted in somewhat lower threshold.

Since an 827 nm ECDL was also available, we also employed it as an OPO idler seed source. The seeding effects are shown in figure 4.15. These results are essentially identical to those present in figure 4.14. Both signal and idler beams are narrowed significantly from an unseeded line width (FWHM) of ~1 nm, to a seed linewidth less than the limit of the spectrometer resolution (~1 cm\(^{-1}\)).

B. Etalon Trace

With 780 nm seeding we have also performed some preliminary measurements using a 2 GHz free spectral range confocal spectrum analyzer, which while not designed for line width measurement of pulsed lasers can, nonetheless, provide a reasonable estimate. The spectrum analyzer was scanned slowly using a computer generated voltage ramp applied to the controller’s external ramp input. An ordinary PIN photodiode captured the transmitted signal which was processed with a boxcar averager and digitized with the computer. A narrow pass band 780 nm filter was placed directly in front of the detector to avoid stray light at 532 or 355 nm.
Figure 4.16: Etalon trace of cw seed laser (upper) and injection-seeded OPO idler output (lower) at 780 nm. Linewidth of seeded OPO output is 200 – 300 MHz, based on 2.0 GHz FSR of etalon.

Figure 4.16 shows typical traces of the cw seed laser (upper) and time averaged pulsed OPO signal output (lower). While the OPO signal output traces are admittedly somewhat non-steady, due to a combination of cavity mode stability and drift in the non-
stabilized etalon, it is clear that the average line width is a fraction (10 – 15%) of the etalon FSR. We conclude that the time averaged line width is on the order of 200 – 300 MHz. While it is somewhat speculative, it should be noted that the longitudinal mode spacing for our OPO cavity is ~ 1.5 GHz. It is possible, although not conclusively determined, that the weak “peaks” which seem to periodically occur at a position ~ one third of the way between the major transmission peaks could be an indication of lasing on an additional longitudinal mode.

4.3.2. Injection Seeding for 532 nm Pumping OPO

The schematic diagram for injection seeding of a 532 nm pumping OPO system is shown in figure 4.11. Since the OPO “signal” output is at 780 nm, we use the 780 nm ECDL as the “signal” seeding source.

Figure 4.17 shows the effect of seeding on the OPO signal output at 780 nm. Note that in this case, unlike for 355 nm pumping, the high reflector of the OPO cavity also reflects the seeding laser beam. Although it is an SRO system, an additional HR is not necessary to achieve substantial linewidth reduction. The seeding effect shown in figure 4.17 is even more significant than that of 355 nm pumping OPO. The linewidth is changed from ~3 nm to the limit of the spectrometer resolution with injection seeding. We haven’t yet performed etalon measurements for this seeding case because obtaining 780 nm OPO output is not our primary goal.
Figure 4.17: Low resolution spectra of seeded and unseeded OPO output using 532 nm pumping.

We also measured the small signal gain of the 532 nm pumping SRO system by using an OMA V system. The 20% output coupler is removed from the OPO cavity to measure the single pass gain. First we measured the 780 nm seeding laser spectrum, which showed a peak value of ~600 arbitrary units. Second, we measured the OPO signal output spectrum at 780 nm with injection seeding, which showed a peak value of ~40000 units with the 532 nm pumping pulse energy of ~34mJ/pulse. Therefore, the single pass small signal gain is ~67 in this case. For a lower pump-pulse energy, 22mJ/pulse, the measured single pass gain is ~13.
4.3.3. Some Comments on Injection Seeding

A. Seeding Effect of an SRO system

When we performed injection seeding of our OPO system, we first tried to seed a 355 nm pumped SRO system. A 780 nm diode laser was used to seed the OPO idler wave, when the OPO cavity was resonant only for the signal wave (at 651 nm). Figure 4.18 shows the OPO signal and idler output spectra with seeding and without seeding. It is very clear that there is no seeding effect in figure 4.18, e.g., idler seeding did not work for a signal resonant (only) OPO cavity. The two OPO cavity mirrors, high reflector and output coupler, have very low reflectance at the idler wavelength of 780 nm. Therefore, the seeding laser beam experiences only a single pass through the OPO cavity. During the course of our experiments, we found that at least two passes were needed to make the injection seeding work, or in other words it was necessary for the high reflector of the OPO cavity to have significant reflectance not only at the signal wavelength, but also at the idler wavelength. In other words, a DRO system is necessary for idler injection seeding.
Figure 4.18: Low resolution spectra of seeded and unseeded SRO output using 355 nm pumping.
B. Some Comments on Injection Seeding for a DRO system

For any optical resonator, such as the OPO cavity shown in figure 4.1, the transmission of the resonator as a function of wavelength is given by\textsuperscript{[23,34]}:

\[
T = \left[ 1 + \frac{4r}{(1-r)^2} \sin^2 \left( \frac{2\pi L}{\lambda} \right) \right]^{-l}
\]  

(4.4)

where \(L\) is the cavity length, \(\lambda\) is the incident wavelength, and \(r\) is the reflectivity given by:

\[
r = \sqrt{R_1R_2}
\]  

(4.5)

where \(R_1\) and \(R_2\) are the reflectance of the cavity mirrors. The maximum value of the transmission occurs when the mirror separation is an integer’s multiple of half the wavelength:

\[
L = \frac{m\lambda}{2}; \ m=1,2,3,...
\]  

(4.6)

Figure 4.19 illustrates the transmission properties of an optical resonator. We define the ratio of the spacing between two adjacent modes, \(\Delta \nu\), and the mode width, \(\delta \nu\), (FWHM) as the finesse:

\[
F = \frac{\Delta \nu}{\delta \nu} = \frac{\pi}{2} \left[ \frac{4r}{(1-r)^2} \right]^{-1/2}
\]  

(4.7)
where the longitudinal mode spacing, $\Delta\nu$, is given by:

$$\Delta\nu = \frac{c}{2L}$$  \hspace{1cm} (4.8)

Figure 4.19: Typical transmission spectra of an optical resonator.

The cavity length of the 355 nm pumping DRO system, which is shown in figure 4.1, is ~10cm, corresponding to a mode spacing of ~1.5 GHz. The linewidth of the ECDL is ~1MHz, which is much narrower than the mode spacing. To perform the injection seeding, one of the cavity mirrors is usually mounted on a piezoelectric transducer for optimization and stabilization of the length of the cavity $^{[70, 81]}$. In this manner, the seed wavelength can be matched to a longitudinal mode. However, in our experiments,
although a piezoelectric transducer controlled cavity mirror was not employed, we still observed very significant seeding effects. There are two reasons for this. First, the output coupler in the OPO cavity has a reflectance of only ~20%, so based on equations (4.5) and (4.7), the finesse is quite low, ~3.8. The corresponding cavity transmission, based on equation (4.4), is shown in figure 4.20.

![Transmission spectra of DRO cavity.](image)

Figure 4.20: Transmission spectra of DRO cavity.
From figure 4.20, we see that even at the minimum point, the cavity transmission is still ~15%. Since the round trip gain of the cavity is very high (>10^3 based on our 532 nm pump measurement), it is clearly still possible to achieve “lasing” amplification, even “off” longitudinal cavity mode resonance. Of course the gain will be lower than that for the idler wave precisely adjusted to the longitudinal mode resonance.

Second, the linewidth of the 355nm pumping beam is quite wide compared to the linewidth of the seeding laser. While the instrumentation was not available to measure the linewidth of the 355 nm pumping laser beam, we can estimate it based on previous work by Lempert et al. [7], who reported a linewidth (FWHM) of ~100MHz at 1064 nm. In addition the system reported by Lempert et al. does not utilize a PCM, which will increase the linewidth, as will be discussed in more detail in chapter 6. Therefore, the linewidth of our fundamental beam is most likely on the order of ~500MHz [85]. After second and third harmonic generation, the linewidth will further increase, resulting in a value close to (or perhaps more than) the OPO longitudinal cavity mode spacing of 1.5 GHz. Therefore, considering both the high gain and the pump linewidth, we conclude that the OPO cavity stabilization for injection seeding is not necessary.
CHAPTER 5

NO BASED PLANAR LASER INDUCED FLUORESCENCE

In the last chapter we presented burst sequences obtained from third harmonic pumping of an OPO. In this chapter we will describe the generation of the NO excitation wavelength, 226 nm, by mixing the OPO “signal” output and the residual third harmonic wave. We will use such 226 nm burst sequences to demonstrate, for the first time by any method, ultrahigh repetition rate NO PLIF imaging.

5.1 Generation of 226 nm

To obtain 226 nm we need to mix the OPO 622 nm output with the residual third harmonic wave. However, the OPO output has some delay time compared to the 355 nm pumping, as is shown in figure 5.1. According to figure 5.1, the pulse width of the 355 nm pumping beam is ~8ns, whereas the OPO signal output has a pulse width of ~6ns, The OPO signal output is also delayed by ~4-6 nsec. Since the OPO is an oscillator, it takes some time to build up a high intensity beam, by means of multiple passes of initial “noise” OPO photons through the cavity. In addition, since OPO optical gain is highly non-linear with pump beam intensity, the output pulse width is “gain narrowed”.
In order to obtain maximum conversion efficiency for 355nm and 622nm mixing, we need to make these two beams not only matched in space, but also in time. We use an optical delay line of ~4 ns, as shown in figure 5.2. The residual 355nm beam transmitted through the OPO cavity is reflected by several mirrors and travels about 4 feet more than the 622 nm OPO output beam. The mirror M1 is a dichroic mirror, which reflects 622 nm and transmits 355nm. (Note that there are some additional optics used, which for simplicity are not shown in figure 5.2, such as the optics for injection seeding and separation of “signal” and “idler” beams.) The mixing crystal is also a Type I BBO crystal, which is 12-mm-long and 8-mm × 8-mm in cross section.
Figure 5.2: Experimental setup for 355 nm and OPO output mixing.

From figure 5.2, we see that the OPO system is used in a single pass pump geometry for frequency mixing. We also explored double pass pumping, but in this case the residual 355nm laser beam is too weak to obtain good mixing conversion efficiency. The reason is the many optical energy losses in the OPO cavity. In particular, the surfaces of the nonlinear crystals used in the OPO cavity are not anti-reflection coated because the coating cannot survive the high intensity. The 355nm pump energy loss is ~33% in a
single pass, so for a double pass pump OPO, two thirds of the pump energy will be lost, which is too much for our experiment.

Figure 5.3 shows a representative burst sequence of 6 pulses with 10 microseconds spacing obtained using single pass pump in the geometry shown in figure 5.2. The average single pulse energy is ~200µJ when we operate the OPO without injection seeding. With 827 nm idler seeding, the mixing conversion efficiency is higher by a factor of 2. Because the pulse energy is very low, and fluctuates considerably, it is somewhat difficult to measure, and the reported value should be considered approximate.

![Figure 5.3: Typical single 6-pulse, 10 us spacing 226 nm burst sequence. Average single pulse energy is ~200 µJ.](image)
The conversion efficiency for 355nm and 622 nm mixing is somewhat lower than what we expected. Before mixing the 355nm and 622 nm pulse energies are \(~11\text{mJ/pulse}\) and \(~4\text{ mJ/pulse}\), respectively, and the 226 nm output pulse energy is only \(~200\text{mJ/pulse}\), which corresponds to a conversion efficiency of \(~3\%\). Therefore, we tried another way to obtain 226 nm. We built a DRO system and made its signal output at 452 nm. The second harmonic of 452 nm is also 226 nm. In this experiment, we used the same BBO crystals for OPO operation, but we changed some mirrors to make a 452 nm resonant DRO cavity. In this case, the phase-matching angle has about 4-degree difference from the cutting angle of the crystals, so the OPO conversion efficiency is a little bit lower. We used the same BBO mixing crystal as the crystal for 452 nm frequency doubling. In the end, we obtained about the same 226 nm energy, \(~200\text{µJ}\) at the same experimental condition.

5.2 NO PLIF Experiment

5.2.1. Experiment Setup

Figure 5.4 shows the experimental apparatus for the NO PLIF proof-of-concept measurement. The 226 nm beam is focused with a cylindrical lens to make a laser sheet and transmitted to a flow chamber. The flow system is similar to that used previously in Molecular Tagging Velocimetry (MTV)\[^{73}\] studies of flows from micro nozzles. Nitrogen seeded nitric oxide is the test gas for a Mach 2 two-dimensional micro nozzle of dimensions 1 x 5mm. The chamber is sealed with quartz windows to transmit UV light.
We use a Princeton Scientific Instruments ultra-high frame rate camera, which was also used in previous ultra-high frame rate Planar Doppler Velocimetry (PDV) experiments\cite{15}, to capture the UV fluorescence.

![Diagram of the camera's architecture](image)

**Figure 5.4:** Experiment Setup of NO PLIF measurement.

5.2.2. PSI-IV Camera

A. The Diagram of the Camera’s Architecture

The camera was manufactured by Princeton Scientific Instruments. and has the ability to capture a sequence of up to 28 images at up to 1 million frames per second (1 MHz). A simplified diagram of the architecture of four pixels of a camera very similar to ours is shown in figure 5.5. For this camera, each pixel consists of a photoactive region
and a 4 x 7 array of memory elements. High frame rates are achieved by shifting charge produced on the active area of the chip to the array of individual memory modules at transfer times as fast as 1 microsecond. Upon receipt of a master trigger, the camera operates continuously until a stop trigger is issued. At this point, the last 28 images are read-out to a computer. In order to preserve a reasonable fill factor, and due to the short exposure times associated with high-speed imaging, the overall size of each pixel is quite large (~115 x 115 microns). The memory storage occupies more than half of this space, resulting in a fill factor for the chip of 47.6%, yielding an effective active pixel size of ~79 microns.

Figure 5.5: Architecture of 2×2 array of pixels from PSI-IV camera.
B. Quantum Efficiency Measurement of PSI-IV Camera in Ultra UV

The quantum efficiency of the PSI-IV sensor had previously been measured by PSI in the wavelength range between 250 and 940 nm. Figure 5.6 shows that data, where the minimum wavelength is 250 nm. However, referring to the NO fluorescence spectrum, shown in figure 1.8 we find that NO fluorescence occurs in the range from 220nm to 300 nm. Since no data was available quantifying the camera’s sensitivity below 250nm, it was important to measure the quantum efficiency in the range 220-250 nm.

Figure 5.6: Quantum efficiency of PSI-IV Imager (>250nm).
The quantum efficiency measurement was accomplished using three narrow spectral band pass transmission filters, centered at 254nm, 224nm and 200 nm, respectively, in combination with a deuterium lamp with output in the range from 110 to 315 nm. First the deuterium lamp intensity was calibrated in this wavelength range using an OMA V system. Then using the lamp as a calibrated light source, the quantum efficiency of PSI-IV imager was measured.

![Diagram](image)

**Figure 5.7**: Deuterium lamp intensity calibration setup.

Figure 5.7 shows the experiment setup for calibrating the deuterium lamp intensity. Each filter was placed in front of the spectrometer slit and the corresponding lamp spectrum was measured. By integrating each spectrum, we obtained relative intensities for three different bands. The relative intensity ratio of the lamp, measured after transmission by the three filters is 200nm:224nm:254nm=1:1.8:3.3. The quantum efficiency of our ICCD camera is ~ constant, ~18% in this 200-250nm range, but the spectrometer grating efficiency varies somewhat, approximately 200nm:224nm:254nm =
1:1.16:1.32, according to manufacturer specification. Taking into account the spectrometer wavelength dependence, we obtained a corrected relative intensity ratio of the lamp as 200nm:224nm:254nm=1:1.55:2.50 after transmission through the respective filters.

![Diagram](Diagram.png)

Figure 5.8: Experiment setup for calibrating QE of PSI-IV camera.

This result was then used to calibrate the quantum efficiency of the PSI-IV sensor. Figure 5.8 shows the experiment setup, which is similar to figure 5.7, except that the PSI-IV camera is substituted for the OMA system. For each filter, we obtained ~100 images with the PSI-IV camera. By analyzing the images, we obtained average intensities, with the result that the ratio of the average intensities for the three filters are 200nm:224nm:254nm=0.45:0.83:1. Considering the light source intensity that was measured earlier, the quantum efficiency ratio of the PSI-IV camera is 200nm:224nm:254nm=1.14:1.33:1. From figure 5.6 we find that the QE is ~34% at 254 nm (as determined by PSI), so the QE at 200nm and 224nm are 39% and 45% respectively. We note that this procedure is rather approximate and it is possible, that the
apparent increase in quantum efficiency with decreasing $\lambda$ below 250 nm is actually due to some systematic error. Nonetheless it is clear that the PSI-IV sensor has substantial quantum efficiency below 250 nm, verification of which was the real goal of these measurements.

5.3 Mach 2 1 x 5mm Micro Nozzle

Figure 5.9 shows a photograph of the body of the nozzle, which was fabricated using the Electric Discharge Machining (EDM) technique. Following an approach described by Bayt and Breuer [74], aluminum walls were then bonded to the top and bottom.

Figure 5.9: Photograph of two-dimensional Mach 2 nozzle. Exit dimensions are 1 x 5 mm.
The facility pumping speed is currently limited by the available pump. The maximum pumping speed is ~10 liter/minute for our pump. We can increase the chamber ambient pressure by partial closing of a valve located between the exit of the dump tank and the pump, but we cannot decrease it. Therefore, the nozzle could be only operated in pressure matched or overexpanded flow regimes.

Note that the flow Mach number at the nozzle exit can be lower than the design Mach number, in some cases, because of viscous effects \cite{75, 76, 77}, even though the pumping speed is otherwise sufficiently high. In previous MTV experiments \cite{73}, we evacuated the system with a larger, 60 liter/minute, pump. Figure 5.10 shows a plot of the measured ratio of stagnation pressure to nozzle exit pressure as function of stagnation pressure when the facility is operated at its maximum, 60 liter/minute, pumping speed.

![Figure 5.10](image_url)

Figure 5.10: Experimental ratio of stagnation pressure to exit pressure as a function of stagnation pressure for 1 x 5 mm nominally Mach 2 rectangular nozzle.
It can be seen that the pressure ratio increases rapidly as the stagnation pressure is increased from approximately 10 to 50 torr, after which it continues to rise more slowly, leveling off to a ratio of ~7.1. This corresponds, assuming isentropic flow at $T_0 = 300$K, to a Mach number of 1.94. Note that the nozzle was designed for Mach 2 using the method of characteristics. Therefore, at higher pressures (stagnation pressure of the order of atmospheric and higher), the measured Mach number is close to the design Mach number and thus viscous effects within the nozzle are relatively small. However, if the stagnation pressure is in the range 10-50 torr, the flow mach number is only ~1.1-1.3. Since the pumping speed was high enough in the previous MTV experiment, by suitable combination of stagnation pressure, flow rate, and pumping speed, the nozzle still could be operated in flow regimes ranging from pressure matched ($P_{exit} = P_{ambient}$) to highly underexpanded ($P_{exit} \sim 5$-$10 P_{ambient}$) in this low stagnation pressure range.

Although the flow Mach number is limited by the pumping speed, we still were able to perform some proof-of-concept high repetition rate NO PLIF experiments by using this facility. The results are shown in the next section.

5.4 NO PLIF Images

After verifying that the PSI-IV camera has good sensitivity in the range corresponding to NO UV fluorescence, we performed some proof-of-concept ultra-high frame rate NO PLIF imaging measurements, using the Mach 2 microjet shown in figure 5.9.
First, however, we consider a simple calculation to estimate the required laser power in our experiments. If we seed 1/4 torr NO into the cell, the NO number density is \(8.8 \times 10^{15}/\text{cm}^3\). The saturation spectral irradiance is defined by equations (1.4) and (1.5). If we assume that collisional quenching and stimulated emission are negligible (at our static pressure and laser beam intensity conditions), based on equation (1.4), the saturation spectral irradiance is \(\sim 8.7 \times 10^{-5} \text{mJ/cm}^2\). If we use a laser sheet of 2 cm long \(\times\) 0.5 mm thick with an assumed linewidth (FWHM) of 1 GHz at 226 nm, and a 6 ns laser pulse duration, the saturation energy is \(\sim 5.2 \times 10^{-5} \text{mJ/pulse}\), which is a very small number.

5.4.1. PLIF Images Based on a 5mm Nozzle Exit

Some initial results, using a gas mixture of 3% NO in pure N\(_2\), are shown in figure 5.11. In this case, the nozzle exit height is 5mm and the flow is from left to right. The image field-of-view is \(\sim\)10x20 mm. The nozzle was operated in an off-design flow regime, 110 torr stagnation pressure and 30 torr ambient pressure. The measured nozzle exit pressure is 30 torr. The measured flow region is \(\sim\)4H-8H from the nozzle exit, where H is the vertical size of the nozzle exit 5 mm. The OPO is unseeded and the image sequence is \(\begin{pmatrix} 1 & 2 \\ 3 & 4 \\ 5 & 6 \end{pmatrix}\) in figure 5.11.
Figure 5.11: NO PLIF images based on a 5mm exit Mach 2 nozzle. The 226 nm energy is \( \sim 150 \, \mu\text{J/pulse} \).

A simple theoretical calculation based on the measured stagnation pressure and the chamber pressure indicates that the jet was overexpanded, which would therefore exhibit an expansion and compression cell pattern. The NO concentration would be high in the compression regions (higher PLIF signal) and low in the expansion regions. The images show such bright and dark regions.
Figure 5.12: NO PLIF images based on a 5mm exit Mach 2 nozzle. The 226 nm energy is ~180 µJ/pulse.

Figure 5.12 is very similar to figure 5.9. The only difference is that the laser pulses have a slightly higher energy (~180 µJ/pulse) than in figure 5.11. The expansion and compression cell pattern is also observed in this figure.

5.4.2. PLIF Images Based on a 1mm Nozzle Exit

As a second example, we rotated the nozzle by 90° so that the nozzle exit height is now 1 mm. The gas mixture is the same as before, 3% NO in pure N₂. The nozzle was still operated in an overexpanded flow regime, but with a higher mass flow rate, accomplished by increasing the stagnation pressure to 248 torr. The measured ambient
(or back) pressure and the measured nozzle exit pressure were both 95 torr. Some PLIF images are shown in figure 5.13.

The flow region is ~0-10H from the nozzle exit, where H is the vertical size of the nozzle exit 1 mm. Each image has ~6 x 10 mm field of view. The image sequence is \( \begin{pmatrix} 1 & 2 \\ 3 & 4 \\ 5 & 6 \end{pmatrix} \) in figure 5.13. The OPO is seeded in this case and the burst sequence contains 6 pulses with 10 µs interpulse spacing. The 226 nm laser pulse energy is ~200µJ/pulse. Again, figure 5.13 shows some bright and dark regions indicating NO concentration differences. However, the images show very coherent asymmetric large-scale structure. This type of behavior would result if the jet was operating in a flapping mode.

Figure 5.13: NO PLIF image sequence obtained from 226 nm burst sequence similar to that shown in figure 5.3. Flow is from left to right, Mach 2 micro nozzle with 6 x 10 mm Field of View.
Since the only flow condition that we can run with our current facility is overexpanded, we also operated the nozzle in several unsteady conditions by continuously changing the stagnation pressure. Figure 5.14, 5.15, and 5.16 show some typical NO PLIF image sequences with unsteady flow conditions.
Figure 5.14: NO PLIF image sequence with unsteady flow.
Figure 5.15: NO PLIF image sequence with unsteady flow.
Figure 5.16: NO PLIF image sequence with unsteady flow.
5.5 Conclusions

These PLIF images demonstrate the ability, for the first time, to perform NO based PLIF (or any similar UV PLIF) imaging at ultra-high repetition frame rates. Other than this experiment, the only other similar imaging capability has relied on 532 nm based Mie scattering and Rayleigh scattering \[6-10,13,15\].

Although the flow facility is admittedly rudimentary and the flow is not of real physical interest, it demonstrates the ability of the technique to be used in high speed and high repetition rate NO PLIF imaging. Also, the static conditions are similar to a Mach 5 facility, located in the U.S. Air Force Research Laboratory, a sponsor of this work.

Finally, we note that improvement in the technique are on-going and we plan to perform additional measurements using an Intensified CCD camera which will be delivered to OSU for testing this summer. As will be described in Chapter 7, we plan to incorporate a larger vacuum pump for those measurements, soon.
CHAPTER 6

SOME RECENT PROGRESS IN PULSE BURST LASER DEVELOPMENT

Since the pulse burst laser is the most critical element of our ultra-high frame rate PLIF and PDV diagnostics, the effort to build a lower price, higher power, and more stable pulse burst laser system is on-going. This chapter will describe some additional progress that have been made during the time period in which the bulk of this thesis was written.

We reported in previous chapters that our pulse burst laser now works quite well. Specifically, it is now possible to obtain reasonably uniform burst sequences of ~6-12 pulses and 4-12 microseconds interpulse spacing with sufficient individual pulse energy to perform NO PLIF and PDV velocimetry. Nonetheless, considerable progress can still be envisioned. The first thing that we need to consider is price, which is rather high (~$200K-$250K) for the current OSU system, and which limits the affordability for many researchers who might otherwise want to use such a laser. The second thing is the pulse energy. The individual pulse energy is in the range of 100-250 mJ/pulse at the wavelength of 1064 nm. While for NO PLIF experiments the 226 nm energy, while somewhat low, is sufficient, some other researchers may need higher energy for their particular applications, an example of which will be discussed later. The third one is the
pulse burst duration. Most burst sequences reported in this thesis have total burst train durations in the range of 60 µs - 100 µs. The time duration is currently limited by the power supplies of our amplifier chain. The current pulsed flashlamp drivers were purchased from Analog Modules Inc. and are similar to those used in most commercial, 10 Hz, lasers. As shown in figure 3.5, this kind of power supply can only support a gain curve of ~300 µs FWHM.

Recent progress in these three areas will be discussed in more detail in the following sections.

6.1 New Pulsed Flashlamp Driver

Recently Analog Modules Inc. has developed a new power supply for pulsed flashlamp driving. The new Model 8800V laser flashlamp controller provides variable pulsewidth pulses for pumping solid-state lasers. The pulse width is variable in the range of 300 µs – 2 ms. We were able to purchase one such controller and measure the double pass small signal gain curve from the first of our amplifiers. The results are indicated in figure 6.1.

If the same total flashlamp energy and pulse width is used as was used with the old lamp driver (~300 µs and 40 J), the Gaussian small signal gain profiles, as shown in figure 6.1, are very similar. Note that the double pass gain is ~1000 with the old power supply. However, we could increase this gain to up to ~2000 with the new power supply, as shown in the left third and fourth curves in figure 6.1. More importantly, with longer flashlamp pulses, in the range of 1ms-2ms, the small signal gain exhibits a flat top profile.
with time duration in the range 300 µs to 1500 µs. This flat top is very important for operation of the pulse burst laser. If we utilize burst sequences in this range, the burst envelope will be very uniform, since every pulse has similar same gain.

Figure 6.1: Gain comparison between old 300 µs and new 2 ms flashlamp controller.

With the new flashlamp controller we were also able to make the total time duration of the burst sequence much longer. If we use a nominal 2 ms duration, the flat top portion of the gain curve is ~1500 µs long. If we were to use a 10 µs interpulse
spacing with this setup, we will have a uniform burst sequence containing 150 pulses. This is a very encouraging result which we plan to exploit in a system to be delivered to the U.S. Air Force Research Laboratory (Propulsion Directorate) within the next 4-6 months.

6.2 Pulsed Master Oscillator

Since the pulse burst laser can produce high repetition rate and high energy laser pulses, it has generated interest amongst many scientific researchers. Dr. Daniel Den Hartog, a senior research staff member in the plasma physics group, Department of Physics, at the University of Wisconsin Madison, has indicated interest in applying the burst mode laser system to Thomson Scattering measurements \cite{78}. Such measurements require individual pulse energy as high as ~1 J/pulse at the fundamental wavelength 1064 nm.

In an effort to simplify the system, we explored the use of a new high repetition rate pulsed Nd:YVO₄ laser, which was purchased from Crystal Lasers Inc., for use as the master oscillator, replacing the current CW laser. The repetition rate of this laser is variable from 1 to 250. With this pulsed laser, the dual Pockel cell slicer, which costs ~$60K, is not necessary. Another advantage of using a pulsed oscillator instead of a combination of CW laser and Pockel cells is that the individual pulse energy is higher. For the old system, the CW laser has a power of ~100mW. After being sliced into 10 nsec pulses, the pulse energy is ~1nJ/pulse. However, the new 250 pulsed Nd:YVO₄ laser has pulse energy of ~0.4 µJ/pulse, which is more than 2 orders of magnitude higher.
than the old system. The higher oscillator energy is potentially very significant. Specially, I will show results that indicate that the pulse energy can be increased to 10’s of mJ/pulse with amplification from only one quadruple-pass, or two double-pass, amplifiers. Such energy is sufficient for performing Mie scattering based planar flow imaging experiments, such as PDV.

With the pulsed Nd: YVO$_4$ laser, we measured the double-pass small signal gain by using the first amplifier shown in figure 3.1. Note that the amplifiers used in the system are Nd:YAG rods pumped by flashlamps. The results are very similar to the gain obtained with Nd:YAG laser, as is expected since there is significant overlap in the spectral gain profile. The maximum gain is still ~1000 when we used vanadate as the gain medium.

6.2.1 Burst Sequences with 10 Pulses and 10 µs Interpulse Spacing

The initial burst sequence studied consisted of 10 pulses with 10 µs interpulse spacing. The laser burst frequency is again 5 Hz, e.g., there are 5 bursts per second. The measured average power is ~950 mW after the first two amplifiers, with 40 Joules flashlamp energy input to each amplifier. The corresponding single pulse energy has been increased from ~0.4 µJ to ~20 mJ, output by the pulsed oscillator after only two double-pass amplification stages. The total gain of the first two amplifiers is ~5 x 10$^4$. We also measured the average Amplified Spontaneous Emission (ASE) power in this configuration, which is only ~2mW.

However, Amplified Spontaneous Emission, which as shown in figure 6.2, consists of a quasi-random output of ~1-2 µsec duration “pulses” spaced by ~3-4 µsec,
grows rapidly as additional amplifiers are added. The power measured after 5 amplifiers is \( \sim 14 \text{W} \), but 12W are from ASE. Therefore we had to find a way to reduce ASE. Spatial filtering is a possible choice, but in practice it is difficult to achieve sufficient ASE reduction. Therefore we used the Phase Conjugate Mirror.

The function of the PCM has been discussed in chapter 3, where we located the PCM after the fourth amplifier, as shown in figure 3.1. For this study, we moved the PCM to in front of the fourth amplifier because the laser pulse energy is now higher than that achieved with the CW oscillator and slicer.

![Figure 6.2: Amplified spontaneous emission in the pulse burst laser system.](image-url)
With a 200 mm FL lens, the laser power reflected by the PCM is ~1.1 W for an input power of 1.9 W. The individual pulse energy ratio is ~38mJ:22mJ between the input and output pulses. Note that we did not use the maximum energy for the first three amplifiers in order to prevent dielectric breakdown in the PCM. The flashlamp energies are only 28J, 28J, and 30J for the 1\textsuperscript{st}, 2\textsuperscript{nd}, and 3\textsuperscript{rd} amplifiers respectively. The energy conversion ratio of the PCM is more than 50% in this case.

As discussed earlier, the PCM will decrease the pulse temporal width because Stimulated Brillouin Scattering is a nonlinear phenomena, which only occurs above a certain intensity threshold. The measured pulse duration after the PCM is ~26 ns, whereas the input pulse width is ~36 ns. The comparison of these pulses is shown in figure 6.3.

![Figure 6.3: The pulse width narrowing effect because of PCM.](image)
After amplification by a total chain of 5 amplifiers, the maximum power that we are able to obtain is \(~12.8\) W, which corresponds to an individual pulse energy of \(~250\) mJ/pulse. In addition, due to use of the PCM, the measured ASE power is only \(60\) mW. Figure 6.4 shows a typical burst sequence with 10 pulses and 10 \(\mu\)s interpulse spacing.

Figure 6.4: Typical burst train pumped by pulsed Nd:YVO\(_4\) laser with 10 pulses and 10 \(\mu\)s interpulse spacing. The total energy of the burst sequence is \(~2.5\) J.
The burst envelope shown in figure 6.4 is reasonably uniform in spite of this very high energy condition. The individual pulse energies fluctuate by $\sim \pm 20\%$, for which there are two reasons. First, the pulsed Nd:YVO$_4$ laser has $\sim 5\%-10\%$ fluctuation in pulse energy. When the pulses are amplified by the 5 amplifiers, this fluctuation grows somewhat. Second, the pulsed Nd:YVO$_4$ laser source has a broad linewidth, $\sim 0.2\text{nm}$, which is much broader than 5 of the CW Nd:YAG laser that was used previously. Therefore, the respective spectral irradiance is lower which will result in larger energy fluctuations after the PCM because the SBS gain is inversely proportional to spectral linewidth. In other words, the input energy to the PCM is not greatly above threshold.

Now let us describe the Stimulated Brillouin Scattering in more detail than discussed previously. Brillouin Scattering results from the scattering of light by sound waves. The acoustic waves (acoustic phonons) produce a periodic modulation of the refractive index. Brillouin Scattering occurs when light is diffracted backward on this traveling “grating”, giving rise to frequency shifted Stokes and anti-Stokes components. This process can be stimulated when the interferences of the laser light and the Stokes wave reinforce the acoustic wave through what is known as electrostriction. Electrostriction is a process in which the high electric field of the light wave acts as a spatially periodic force, termed ponderomotive force, on the medium, which produces a periodic modulation in fluid density, which then “launches” a coherent acoustic wave.

In physics, a ponderomotive force is a nonlinear force that a charged particle experiences in a rapidly oscillating, inhomogeneous electric or electromagnetic field. The ponderomotive force $F_p$ is expressed by
\[ F_p = -\frac{e^2}{4m_e\omega^2} \nabla E^2 \]  \hspace{1cm} (6.1)

where \( e \) is the electrical charge of the particle, \( m_e \) is the mass, \( \omega \) is the frequency of oscillation of the field, and \( E \) is the amplitude of the electric or electromagnetic field.

Since the scattered light undergoes a Doppler frequency shift, the Brillouin shift \( v_B \) depends on the acoustic velocity and is given by

\[ v_B = \frac{2nV_a}{\lambda} \]  \hspace{1cm} (6.2)

where \( V_a \) is the acoustic velocity within the medium, \( n \) is the index of refraction and \( \lambda \) is the vacuum wavelength of the incident lightwave. The shape of the Brillouin Gain Spectrum (BGS) is determined by the strong attenuation of sound waves. Actually, the exponential decay of the acoustic waves results in the gain profile, \( g_B(v) \), exhibits a Lorenzian spectral profile \(^{[82, 83]}\). (Note: Fourier transform of an exponential function is a lorentzian.)

\[ g_B(v) = g_0 \frac{(\Delta v_B / 2)^2}{(v - v_B)^2 + (\Delta v_B / 2)^2} \]  \hspace{1cm} (6.3)

where \( \Delta v_B \) is the Brillouin linewidth (FWHM). The BGS peaks at the Brillouin frequency shift \( v_B \), and the peak value is given by the Brillouin gain coefficient \( g_0 \)

\[ g_B(v_B) = g_0 = \frac{\omega_s^2 (\gamma^e)^2}{c^3 n V_a \rho \Delta v_B} \]  \hspace{1cm} (6.4)
where $\omega_s$ is the frequency of the Stokes-shifted Brillouin-scattered light, $c$ is the speed of light, $\rho$ is the medium density. The electrostriction constant is given by

$$\gamma^e = \rho \left( \frac{\partial \varepsilon}{\partial \rho} \right)_T$$  \hspace{1cm} (6.5)

where $\varepsilon$ is the dielectric constant. For a given Brillouin medium and pump wavelength, the gain coefficient depends on the density, the electrostriction constant, and the linewidth.

The Brillouin linewidth in a liquid$^{[84]}$ is given via the relation:

$$\Delta v_B = \frac{\Gamma}{2} \left( \frac{4\pi n}{\lambda} \right)^2$$  \hspace{1cm} (6.6)

and the damping $\Gamma$ by:

$$\Gamma = \frac{1}{\rho} \left( \frac{4}{3} \eta + \eta' + \frac{\kappa}{C_p} (\gamma - 1) \right)^2$$  \hspace{1cm} (6.7)

Here $\gamma$ is the ratio of specific heats $\gamma = C_p / C_v$, $\eta$ is the shear viscosity, $\eta'$ is the bulk viscosity and $\kappa$ is the thermal conductivity of the liquid.

The SBS phenomenon is widely used to create what is known as a Phase Conjugate Mirror. In fact, the SBS reflectivity is nearly equal to that of an ordinary mirror in the narrow band case wherein the pump bandwidth $\Delta v_p$ is smaller than the Brillouin linewidth $\Delta v_B$, which corresponds to a steady-state SBS$^{[85]}$. However, if the laser systems involving the use of an SBS-PCM have a broadband spectrum, it produces a broadband-pumped SBS$^{[86]}$, albeit with a potentially reduced gain.
For a broadband pump, the SBS reflectivity depends on the relationship between four parameters: the coherence length $l_c$ of the laser, the characteristic interaction length $z_0$ which is usually equal to the Rayleigh range, the mode spacing $\Omega_m$, and the Brillouin linewidth $\Delta v_B$. When the coherence length is longer than the interaction length ($l_c > z_0$), the SBS gain for the broadband pump is as high as that for the narrow band pump\[^{86}\]; hence, the SBS-PCM is most likely to apply to a laser system satisfying the condition $l_c > z_0$. The SBS gain was reported to be the same as that for a single longitudinal mode pump and to be independent of the mode structure if the pump laser mode spacing exceeds the Brillouin line-width ($\Omega_m > \Delta v_B$)\[^{87, 88}\].

Reference [85, 86] reported SBS-related properties of fluorinert liquids, which is shown in table 6.1.
<table>
<thead>
<tr>
<th>Fluorinert Property (at 25 °C, 1.06 µm)</th>
<th>FC 72</th>
<th>FC 75</th>
<th>Notes</th>
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<td>Absorption coefficient $\alpha$ (cm$^{-1}$)</td>
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<td>$&lt;10^{-5}$</td>
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<tr>
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<td>563</td>
<td>Directly from frequency shift</td>
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<td>3.4</td>
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<td>Calculated</td>
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<tr>
<td>Stimulated Raman Scattering</td>
<td>Not detected</td>
<td>Not detected</td>
<td>Measured</td>
</tr>
</tbody>
</table>

Table 6.1. SBS-Related Properties of Fluorinert Liquids.

From table 6.1, we see that the Brillouin linewidth $\Delta v_B$ is ~350 MHz for FC-75 with narrow-band laser pumping at 1064 nm, which is much narrower than the linewidth of our Nd:YVO$_4$ laser (~0.2nm).
6.2.2 Burst Sequences with 20 Pulses and 4 µs Interpulse Spacing

As a second example, we used burst sequences consisting of 20 pulses and 4 µs interpulse spacing. After amplification by the first three amplifiers (with flash lamp energies of 30J, 30J, and 35J, respectively), the burst train average power is ~2.4W, which corresponds to a single pulse energy of 24 mJ/pulse. The ASE power is measured to be only 4 mW, which is the lowest that can be achieved without spatial filtering.

![Graph showing pulse width narrowing](image)

Figure 6.5: The pulse width is narrowed from ~75 ns to ~40 ns because of the PCM effect.

After back scattering from the PCM, which is located between the third and fourth amplifiers, the average laser power is reduced to ~0.7 W (7 mJ/pulse). The energy conversion efficiency is only about 29% because the individual pulse energy is lower than the 10 pulse burst case. In addition, the width of the individual pulses created by the
pulsed oscilator is quite long (~75 nsec). The long pulse is a result of the diode-pump laser being operated at very near its lasing threshold when operated at 250\textsuperscript{o}. The diode lasers which pump the Nd:YVO\textsubscript{4} rod have fixed output power, so that the energy stored in the rod is inversely proportional to the time interval between the pulses. However, since the pulse energy is low, the pulse width is decreased substantially by the PCM. Figure 6.5 shows the comparison of the pulse widths before and after the PCM. It can be seen that the original ~75 ns pulse is reduced to ~40 nsec because of the nonlinear nature of the SBS process.

The purpose of using the PCM is to eliminate the ASE. After amplification by a total of 5 amplifiers, two of which are down stream of the PCM, the average laser energy rises to 10.6 J, corresponding to an individual pulse energy of ~106 mJ/pulse at the fundamental wavelength 1064 nm. The ASE power that has been measured is very low, 60 mW, or ~0.06\% of the laser power. A typical burst sequence of 20 pulses with 4 \(\mu\)s interpulse spacing is shown in figure 6.6.

From figure 6.6 we see, however, that the burst envelope is quite non-uniform compared to other burst sequences that have been reported in this dissertation. As discussed in the last section, the combination of the low pulse energy and broad spectral linewidth of the Nd:YVO\textsubscript{4} seeding laser, results in substantial fluctuation in reflection efficiency of the PCM. The resulting burst sequence is less uniform than that obtained with 10 pulses because the oscillator pulse intensity is even lower in this case, resulting in PCM operation very near threshold. Recall that the burst sequence with 10 pulses has an individual pulse energy of 38 mJ/pulse, and temporal width of ~36 ns. We believe that in this 20 pulses case, the PCM input pulse energy/width of 24 mJ/75ns is very close to the
SBS intensity threshold. Therefore, the resulting burst envelope is very nonuniform and the pulse energy also fluctuates greatly, $\sim \pm 40\%-50\%$.

Figure 6.6: Typical 20 pulses and 4 µs interpulse spacing burst train obtained with a PCM. The total energy of the burst sequence is $\sim 2.6$ J.

Figure 6.7 shows a typical burst sequence obtained under the same conditions as those of figure 6.6, except that the PCM has been removed from the system. In this case, the total average laser power is $\sim 17$ W, but the ASE power is $\sim 10.5$ W. Therefore, the laser pulse energy is only $\sim 65$ mJ/pulse. Although the measured ASE is only 4 mW after the third amplifier, it grows up very rapidly in the last two amplifier stages. However, the burst envelope shown in figure 6.7 is much more uniform than that shown in figure 6.6.
because there is no PCM threshold effect. Although it is not as uniform as the burst trains that we obtained with a CW Nd:YAG laser, it is still more uniform than figure 6.6. The energy fluctuation is now about $\pm 30\%$, which grows from the $\pm 5\%-10\%$ fluctuations of the original vanadate laser.

![Graph](image)

Figure 6.7: Typical 20 pulses and 4 $\mu$s interpulse spacing burst train obtained without PCM. The total energy of the burst sequence is $\sim 1.3$ J.

From these rather preliminary results, we conclude that the use of a low energy and high repetition rate pulsed Nd:YAG or Nd:YVO$_4$ laser as the oscillator source in the pulse burst laser system is, potentially, an attractive option. With the pulsed laser source, the Pockel cells are not necessary, and it is likely that at least one stage of amplification
can be removed. However, the effect of ASE needs to be better addressed. As can be seen from the discussions earlier, the Stimulated Brillouin Scattering based Phase Conjugate Mirror greatly benefits from a narrow spectral linewidth oscillator laser. While the Nd:YVO\textsubscript{4} laser purchased by the Wisconsin group is broad band, a line narrowed (~100 MHz) option is available from the manufacturer, and would appear to be a potential solution. Alternatively, or perhaps in combination, spatial filtering may be another good option.

6.2.3 Second Harmonic Generation

As an initial attempt at Second Harmonic Generation we employ a 10-pulse burst sequence with 10 µs interpulse spacing, similar to that of figure 6.4. The individual pulse energy is ~110 mJ/pulse at the fundamental wavelength 1064 nm. Here we didn’t use the maximum pulse energy that we can obtain (~250 mJ/pulse). We tried both Type I and Type II phase-matching by using a KTP crystal and an LBO crystal, respectively. The resulting second harmonic pulse energies are approximately the same in both conditions, ~15mJ/pulse at 532 nm. This corresponds to a conversion efficiency of only ~14%, which is quite low compared to ~50% that we have achieved previously. The most likely reasons for this are two fold. First, the pulse width of ~26 nsec is much longer than the ~10 nsec which results from the pulse sliced system. Second, the laser linewidth is also much broader, because, as discussed above, the pulsed seeding laser does not employ line narrowing. Similar to the SBS effect, discussed earlier, these two effects will result in lower conversion efficiency for second harmonic generation.
6.3 Some Recent Progress at Auburn University

Brian Thurow, a former Ph.D. student at OSU and a co-worker in some of the studies described in this thesis, is building another pulse burst laser system at Auburn University. He is using the new variable pulse width flashlamp controller described earlier and is also incorporating a significant change in the slicer design. In place of dual Pockel Cells (electro-optic slicer) they are using an acousto-optic modulator (AOM).

AOM, also called a Bragg cell, consists of a crystal in which standing acoustic waves are established, which produces a spatial modulation in the index of refraction. The result is a diffraction grating, with line spacing determined by the distance between the standing acoustic wave maxima.

Most AO devices operate in the Bragg regime, where most of the incident laser beam is diffracted into the 1st order with the remaining modes (not including the 0th) annihilated by destructive interferences. This form of diffraction occurs at the Bragg angle, \( \theta_B \), given by:

\[
\sin \theta_B = \frac{\lambda}{2n\lambda_s}
\]  

(6.8)

where \( \lambda \) is the wavelength of light in air, \( \lambda_s \) is the acoustic wavelength and \( n \) is the index-of-refraction. By modulating the intensity of acoustic waves, via a piezoelectric transducer bonded to the crystal, the amplitude of the deflected light can be controlled. Other applications include beam deflection and frequency shifting, where the acoustic frequency is varied as opposed to the intensity.
Pulse slicing can be done by rapidly turning the deflection ‘on’ or ‘off’ via control of the acoustic waves inside the AO crystal. In this case, the deflected laser beam constitutes the desired burst of pulses with the beam only being deflected when a positive voltage is applied to the AOM driver. Since the interested pulse duration is \(~10\) nsec, the most important properties are the rise/fall time. The rise time, \(t_r\), of the deflected pulse is primarily limited by the time it takes for the acoustic wave to travel across the aperture of the beam and is given by:

\[
t_r = \frac{D}{V_a}
\]  

(6.9)

where \(D\) is the diameter of the focused beam within the AO medium and \(V_a\) is the acoustic velocity. Thurow et al. reported a rise time of \(~10\text{-}15\) nsec, which is slower than the \(~3\text{-}4\) nsec rise times achieved with Pockels cells. The reported pulse width is \(~20\) nsec (FWHM).

The advantage of the AOM is the lower price (\(~\$6\text{K}\)) compared with the dual Pockel Cells (\(~\$60\text{K}\)). With the AOM as the slicer, Thurow et al. have shown burst sequences with repetition rates up to \(~5\) MHz. However, since the amplifier chain has not been completed, they reported the individual pulse energy (at the fundamental wavelength \(1064\) nm) of on the order of \(~1\text{-}10\) mJ/pulse. Second harmonic generation has not yet been reported.
CHAPTER 7

CONCLUSIONS AND FUTURE WORK

7.1 Conclusions

This thesis has documented the development of a MHz repetition rate pulse burst laser system. By adding a PCM to the system we improved second harmonic efficiency, to as high as 50%, compared to ~6% without the PCM. High conversion efficiency is obtained for both Type I phase matching, with a 20 mm long LBO crystal, and Type II phase matching, with a 12 mm long KTP crystal. Based on this system we also obtained third harmonic (355nm) burst sequences, using two Type I 20mm long LBO crystals. The conversion efficiency for third harmonic can reach as high as 40% of the fundamental power. Some high energy fundamental, second harmonic, and third harmonic burst sequences consisting of 1 – 12 pulses separated in time by between 4 and 12 microseconds are now routinely obtained. In a 6-pulse, 10 µs interpulse spacing burst sequence, the average individual pulse energy is 130 mJ at 1064 nm, 66 mJ at 532 nm and 52 mJ at 355nm. The reported burst envelopes are quite uniform.

By using Mie scattering from a 28-pulse, 4 µs interpulse spacing burst sequence at 532 nm, in collaboration with Brian Thurow and Marc Blohm, two former GDTL
students, we have demonstrated quantitative ultrahigh frame rate velocity imaging, using the Planar Doppler Velocimetry technique. The pulse burst laser is paired with two ultra-high repetition rate (PSI-IV) cameras, which are used to capture the resulting planar image sequences.

We have also demonstrated the ability to generate ultra-high frequency sequences of broadly wavelength tunable, high intensity laser pulses using a home built injection seeded Optical Parametric Oscillator, pumped by the second and third harmonic output of the pulse burst laser. Typical OPO output burst sequences consist of 6 – 10 pulses, separated in time by between 6 and 10 microseconds.

With third harmonic pumping of the OPO system, we studied four conditions, two-crystal SRO cavity, three-crystal SRO cavity, single pass two-crystal DRO cavity and double pass two-crystal DRO cavity. The last two cavities took most of our efforts, where DRO means that both signal and idler waves can circulate within the cavity. The double pass two-crystal DRO cavity gives the best operation in burst mode. For single pass OPO, the average total OPO conversion efficiency is approximately 25%, corresponding to individual 622 nm single pulse energy of ~ 4.8 mJ. For double pass OPO, the average total OPO conversion efficiency is approximately 35%, corresponding to individual 622 nm single pulse energy of ~ 6.2 mJ.

As a preliminary work, we studied 532nm pumping of a single crystal OPO cavity. With single pulse pumping, the conversion efficiency can reach 30%. With multiple pulses pumping, the pulse energy of the OPO output burst sequences is not stable and the burst envelopes were also not uniform.
For both 355nm and 532 nm pumping OPO, we have demonstrated injection seeding. Seeding with external cavity diode seeding lasers at 780nm and 827 nm, the OPO output light linewidth is reduced from 1nm (355nm pumping) and 3nm (532nm pumping) to a value that cannot be measured using our grating spectrometer. Some preliminary etalon traces are also reported. The measured OPO output linewidth is ~ 200-300MHz for both the signal and idler waves with injection seeding.

By mixing the OPO signal output at 622 nm with residual third harmonic at 355 nm, we obtained 226 nm burst sequences with average pulse energy of ~0.2 mJ. Injection seeding of the OPO increases the energy achieved by a factor of ~2. 226 nm burst sequences with reasonably uniform burst envelopes are reported.

Using the system we have obtained, for the first time by any known method, PLIF image sequences at ultrahigh (≥100kHz) frame rates. In particular, we have demonstrated ultra high frame rate NO PLIF imaging, which is particularly difficult due to the required 226 nm deep UV wavelength.

We have also studied the possibility of utilizing a 250 kHz pulsed Nd:YVO₄ laser as the master oscillator. 10-pulse-10-µs spacing burst sequences with reasonably uniform burst envelope have been obtained. The total energy of the burst sequence is ~2.5J.

7.2 Future Work

For further development of the pulse burst laser system, we recommend use of a narrow linewidth high repetition rate pulsed Nd:YVO4 laser as the oscillator, in combination with a PCM and, possibly, a spatial filter (to remove ASE). A high
repetition rate flashlamp pumped, multiple Q-switched laser is also a possible option, although this needs to be studied to determine if it is really viable. The use of an AOM is another possible choice, primarily to the cost, but the relatively long pulse width may be a significant obstacle.

In chapter I the work of Kaminski and Hult et al. [3,4] was discussed, who used a cluster of four individual double-pulsed Nd:YAG lasers to produce 8 pulses in a burst train. Each laser head was fitted with a double pulse option (DPO), where the “Q-Switch” can be pulsed twice in rapid succession during a single firing of the flashlamps. This allows the extraction of up to two laser pulses from each cavity with a selectable time separation of ~100 us. With this system they reported bursts of 8 pulses with individual pulse energies as high as 270 mJ at 532 nm. However, the disadvantage of this approach, in addition to its very high cost (~$300K just for the Nd:YAG itself) is that you cannot obtain more than 8 pulses/burst. The number of pulses, e.g., the repetition rate, is limited by the gain relaxation of the Nd:YAG rod. High repetition rate and high energy cannot be satisfied simultaneously.

Based on this work, we have the idea that if we don’t need extremely high repetition rate (~ MHz), we could use a cavity resonant laser to obtain 10’s or 100’s of kHz repetition rate laser pulses, just by multiple firing of the Q-Switch. For fixed flashlamp power, the laser pulse energy depends on the repetition rate. High pulse energy can be obtained with low repetition rate, and high repetition rate can be obtained with low energy. For example, based on Kaminski’s result, 270 mJ/pulse at 532 nm can be, potentially, obtained with a 10 kHz repetition rate, if the flashlamp power and duration is sufficient. A new Analog Modules Model 8800V flashlamp controller could be applied to
the system to give a 2ms gain time. Then with 10 kHz repetition rate, a 20 pulse burst sequence with ~270 mJ/pulse at 532 nm could potentially be obtained.

Another idea focuses on a high repetition rate, multiply Q-switched oscillator laser, similar to the Nd:YVO$_4$ laser discussed in the previous chapter. Although operation in the MHz regime results in low pulse energy, we could still use such a pulsed laser source as the oscillator at high repetition rate (less than ~15 µs interpulse spacing). It may be possible to increase the reflectance of the output coupler of the laser cavity (therefore reduce threshold) and also make the cavity length as short as possible (to reduce the temporal pulse duration). If even only ~1mJ/pulse can be generated in this manner at repetition rates of ~1 MHz, the total cost of the pulse burst laser system will be significantly decreased.

For OPO operation, it would be useful to study the linewidth more carefully. In particular it would be important to determine the third harmonic pump beam linewidth, which has not performed due to the lack of a suitable, 355nm, etalon. A better etalon for measurement of the signal and idler beam linewidths would also be useful. A 355 nm optical isolator, if one can be purchased, would be useful in order to enable double pass pumping of the OPO for 226 nm mixing purposes. We could also try an OPO signal wavelength of 452 nm and doubling it directly to obtain 226 nm. The 452 nm doubling crystal could potentially be located inside the OPO cavity to increase the conversion efficiency.
Molecular Tagging Velocimetry (MTV) is a "time-of-flight" technique in which a laser is used to "write" a line (or set of lines) into a flow by means of an optical resonance with a suitable target tracer molecule. Upon absorption the tracer molecule exhibits radiative emission, fluorescence (or phosphorescence), in the flows. Velocity is determined by imaging the Laser Induced Fluorescence from the initially excited line a suitable time delay after excitation. The measurement requires only a single pulsed laser for the tagging step, and an intensified CCD camera for the subsequent imaging (or "interrogation") step. Figure 7.1 shows typical MTV images. The right image, obtained 10 nsec after tagging, shows approximate initial tagging position. Flow direction is from right to left. The left image is obtained 400 nsec after tagging. The flow velocity can be obtained by comparing these two images.

Figure 7.1: Typical MTV images. Flow is from right to left.
Building on the process that we have made in high repetition rate NO PLIF imaging, a burst mode NO based MTV technique will be developed to study the correlation of turbulence structures in supersonic flows. Traditionally, single pulse NO based MTV has been studied previously \cite{89} in supersonic flows. NO has a spontaneous radiative lifetime of ~220ns, and delay times in the range of ~500-1000 ns have been reported in hypersonic flows \cite{89}. We plan to extend this technique to ~MHz repetition rates, using the burst mode laser/OPO developed in this research in combination with a new, intensified PSI-IV framing camera, which is currently under development. The ability to gate the intensifier will provide the accurate timing control required for the MTV measurement. We plan proof-of-concept measurements in the Mach 2 facility, which will be equipped with a high speed pump in order to provide better flow quality.


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