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Spectral and temporal characteristics of a subpicosecond KrF excimer laser

Le Blanc, Stephen Paul, M.S.
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SPECTRAL AND TEMPORAL CHARACTERISTICS OF A SUBPICOSECOND KrF EXCIMER LASER

BY

STEPHEN PAUL LE BLANC

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R. Sauerbrey, Associate Professor of Electrical and Computer Engineering, Director

F. K. Tittel, Professor of Electrical and Computer Engineering

N. Halas, Assistant Professor of Electrical and Computer Engineering

HOUSTON, TEXAS
APRIL, 1991
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ABSTRACT

Spectral and temporal characteristics of a subpicosecond KrF excimer laser are determined with the use of a one meter spectrometer, a single shot intensity autocorrelator, and a single shot phase sensitive autocorrelator. Spectral analysis is shown to be a simple and sensitive method for aligning the laser system. Experimental results and model calculations show that the center of the laser spectrum is influenced by self phase modulation. Near the edge of the amplifier gain bandwidth, the spectrum is also influenced by the vibrational levels of the KrF upper state. To support the spectral analysis, temporal measurements are performed with single shot autocorrelators. Using a single shot autocorrelator based on three photon fluorescence of XeF, the pulse width was measure at 477 fs. To determine the frequency chirp of the laser, a single shot phase sensitive autocorrelator for ultraviolet lasers was designed, and preliminary tests were performed.
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Introduction

Recently, several research groups have developed high power, ultrashort pulse laser systems operating in the ultraviolet. These subpicosecond ultraviolet laser systems are based on short pulse visible dye laser systems in which the dye laser output is frequency doubled and then amplified in an excimer gain medium. Colliding pulse mode-locked, distributed feedback, and synchronously pumped dye lasers are the most commonly used dye laser systems for generating subpicosecond visible laser pulses. Excimer amplifiers are used to amplify the frequency doubled subpicosecond ultraviolet laser pulse because these amplifiers have high gain coefficients and sufficiently large gain bandwidths (~100 cm\(^{-1}\)) necessary for short pulse amplification.

High power, ultrashort pulse lasers are capable of producing focal-spot intensities greater than 10\(^{16}\) W/cm\(^2\). At such high intensities, the electric field strength produced by the focused laser pulse is comparable to the atomic electric field strength. These high brightness systems, thus, provide the unique opportunity to study the interaction between light and matter in an intensity regime where non-linear effects are expected to be dominant. To date, high brightness ultraviolet laser systems have been used to observe multiphoton absorption in atoms, create short pulse X-ray radiation [1], study high density laser produced plasmas [2,3], and produce supercontinua extending to the vacuum ultraviolet [4,5]. Through time resolved spectroscopy [6] and reflectivity measurements, these laser systems are also capable resolving transient phenomena occurring on a subpicosecond time scale.

In order to interpret the results in the types of experiments mentioned above, it is essential that the characteristics of the laser system itself be well known. It is especially important to develop simple and meaningful diagnostics for subpicosecond ultraviolet lasers. Short pulse, high energy ultraviolet lasers operate at lower repetition rates (<20 Hz)
than short pulse, low energy visible lasers (~100 MHz). Additionally, there are fewer transparent materials and non-linear optical processes available for short wavelength lasers. The purpose of this thesis is to present some of the temporal and spectral diagnostic procedures that have been developed to characterize a subpicosecond KrF excimer laser system. A brief description of this laser is given in Chapter 1. More detailed information and comprehensive alignment procedures appear in Appendix A. Chapter 2 discusses a simple and sensitive method for tuning and stabilizing the laser by examining the spectral behavior of the laser system. The spectral behavior of the laser system is shown to be dependent on self phase modulation and the vibrational structure of the excimer transition. Methods for determining the temporal duration of the laser pulse are presented in Chapter 3.

The subpicosecond KrF excimer laser system has been designed so that the frequency of the laser pulse varies in time - a phenomenon that is usually called frequency chirping. The laser pulse is appropriately chirped so that the pulse width can be further reduced by passing the pulse through dispersive elements. Ideally, design of the proper dispersive elements requires information about how the frequency varies in time. Unfortunately, neither the spectral diagnostic methods discussed in Chapter 2 nor the pulse width measurements examined in Chapter 3 are sensitive to the functional form of the frequency chirp. Several methods for determining the frequency chirp of subpicosecond visible lasers have been developed. However, these methods can not be used for subpicosecond ultraviolet lasers. In Chapter 4, the development of a method to determine the frequency chirp of subpicosecond ultraviolet lasers is discussed and preliminary results from this method are presented.
References


Chapter 1: Subpicosecond KrF Excimer Laser System

I. Introduction

The subpicosecond excimer laser system that is described in this paper is the result of research into pulse shortening techniques that was conducted by Szatmari and Schafer [1,2,3] at the Max-Planck-Institut fur biophysikalische Chemie and by Simon and Gerhardt [4] at the Laser Laboratorium in Gottingen. Although this laser system can be used to produce subpicosecond pulses in XeCl (λ=308 nm) or KrF (λ=248 nm), the KrF excimer laser system has been shown to yield higher output energies and shorter pulse widths. The subpicosecond KrF excimer laser system consists of a double chamber Lambda Physik EMG 150 excimer laser and a subpicosecond dye laser system (Figure 1.1). One chamber of the excimer laser is filled with a XeCl gas mixture and is used to pump the subpicosecond dye laser system that emits 500 fs pulses at 496 nm. The second harmonic of the green laser pulse is generated in a β-barium borate (BBO) crystal. The frequency doubled laser pulse at 248 nm is then amplified in the other chamber of the excimer laser which is filled with a KrF gas mixture. After one pass through the KrF excimer amplifier, the laser beam is spatially filtered to suppress amplified spontaneous emission (ASE), expanded to fill the cross section of the excimer laser chamber, and sent through the KrF excimer amplifier again. The ultraviolet laser pulse emerging from the second pass though the amplifier has a peak power of 25 GW, and the entire laser system operates with a repetition rate between 1 and 5 Hz.

The subpicosecond KrF excimer laser pulse is generated by pulse shaping and pulse shortening mechanisms in a visible dye laser system. The dye laser system consists of a series of dye lasers and amplifiers that can be divided into three sections: a picosecond dye laser, a distributed feedback dye laser, and a two stage amplifier (Figure 1.1). Because the performance of the entire laser system is governed by the the interrelationship among each
Figure 1.1 Block diagram of the subpicosecond KrF excimer laser system. One chamber of a Lambda Physik EMG 150 excimer laser is filled with XeCl and pumps a subpicosecond visible dye laser. The visible pulse is frequency doubled with the BBO crystal. The resulting ultraviolet pulse is amplified twice in the other chamber of the EMG 150. The final 10 mJ output pulse at 248 nm is 500 fs in duration.
of the three sections, it is important to understand how each section works. This knowledge is used later to interpret the spectral and temporal diagnostics that are discussed in Chapters 2, 3, and 4.

II. Picosecond Dye Laser

The picosecond dye laser section consists of a sequence of dye lasers that produce pulses with successively shorter pulse duration. The purpose this section of the laser system is to provide a short (~10 ps) pump pulse for the distributed feedback dye laser that is described in the next section. The picosecond dye laser section consists of a quenched cavity dye laser (QCDL), a short cavity dye laser (SCDL), and two amplifiers separated by a gated saturable absorber (GSA).

The operation of a quenched cavity dye laser can be explained with the aid of Figure 1.2. In Figure 1.2a, a quenched cavity dye laser is formed by two resonators surrounding a common active medium [4,6]. The larger of the two resonators has a longer build-up time for lasing and better amplification per pass. Alternatively, the longer resonator has a high Q value and the small resonator has a low Q value. When the dye is pumped, the shorter resonator lases first with the sharp rise-time characteristic of dye lasers. However, after the build-up time of the longer resonator is reached, lasing in the active medium switches from the shorter to the longer resonator. In this manner, a ~500 ps pulse can be obtained from the short, or quenched, cavity laser when it is pumped by a ~20 ns XeCl laser pulse. The QCDL in the picosecond dye laser section consists of a 5 x 5 x 20 mm³ dye cell filled with a 4 x 10⁻³ molar (M) solution of p-terphenyl in cyclohexane. The quenched cavity is formed by the side walls of the dye cell and the higher Q cavity is formed by an external aluminum mirror and the dye cell walls (Figure 1.2b). With this set-up, the QCDL generates ~400 ps [4] pulses at 340 nm that are used to longitudinally pump the short cavity dye laser.
Figure 1.2 a) Schematic diagram of a quenched dye laser. Lasing in the short, low Q resonator is terminated by more favorable conditions present later in the longer, high Q resonator. The quenched dye laser pulse has a pulse width of less than 1 ns and is spatially separated from the laser pulse from the high Q resonator. b) Schematic diagram of the QCDL. The low Q cavity is formed by the dye cell walls and the high Q cavity is formed by the external mirror M3 and the dye cell walls.
The short cavity dye laser is used to produce a laser pulse with a pulse duration that is roughly a factor of ten smaller than its pump pulse. The cavity is made up of a back mirror that is 60% transmissive at the pump wavelength and 100% reflective at the lasing wavelength and an output coupler that is 80% reflective at the lasing wavelength. Filled with $1 \times 10^{-3}$ M solution of BPBD in ethanol, the dye laser cavity is 500 $\mu$m long. The pulse width of the SC_DL is approximately two to three times the photon lifetime in the cavity, or about 18 ps [6,7]. To achieve the shortest pulse, the SC_DL is pumped approximately five times its threshold for lasing. As the the pumping of the SC_DL increases from threshold to five times threshold, the pulse duration of the SC_DL decreases due to the fast rise and fall times of the dye in the small cavity [8]. Increasing the pumping above five times threshold causes a tail to develop in the pulse shape; this trailing component matches the trailing component of the pump pulse causing the SC_DL pulse to be lengthened. The 365 nm pulse emerging from the SC_DL is then amplified by two saturated amplifiers separated by a gated saturable absorber.

Acting as another pulse shortening device, the gated saturable absorber reduces the pulse width of the SC_DL to 8 ps [1]. The GSA consists of a $1 \times 10^{-3}$ M solution of BBOT in methanol in a 1 mm resonator formed by dichroic mirrors. The mirrors are 10% reflective at the input laser pulse wavelength and 100% reflective at the lasing wavelength of the dye. Saturated absorption and stimulated emission from the dye solution define a time window during which the GSA is transparent to the input laser pulse [6]. Initially opaque to the incident laser pulse, the GSA becomes transparent when absorption of the incident laser pulse saturates (a phenomenon known as bleaching). The GSA becomes opaque again when the build up time of the resonator is reached and the dye begins to lase. When the dye begins to lase, stimulated emission in the dye depopulates the upper absorber state which again increases absorption of the input pulse [9]. By proper design of the resonator, the stimulated emission in the dye is delayed with respect to the bleaching of the
dye and the central and trailing portions of the input pulse are reduced. Since the time window during which the GSA is transparent is narrower than the incident laser pulse width, the transmitted laser pulse width is reduced. The combined effects of the QCDL, SCDL, and the GSA produce an 8 ps dye laser pulse at 365 nm that is used to pump a distributed feedback dye laser.

III. Distributed Feedback Dye Laser

Unlike most lasers, the distributed feedback dye laser (DFDL) has no external resonator for feedback. Feedback in a DFDL is provided by periodic perturbation in the optical characteristics of the gain medium. Operable over a wide spectral range, distributed feedback dye lasers have proven to be simple and inexpensive sources for ultrashort dye laser pulses.

The primary requirement for a DFDL is the formation of high visibility interference fringes in a dye solution [2]. One example of a set-up satisfying this requirement is shown in Figure 1.3a. The pump beam passes through a cylindrical lens and is perpendicularly incident on a grating with a groove spacing d. The +1 and -1 order diffracted beams are internally reflected in a quartz block and focused in a dye cell. The interference fringes formed by the two intersecting beams have a fringe period \( \Lambda \) given by:

\[
\Lambda = \frac{\lambda_p}{2\sin\theta}
\]

where \( \lambda_p \) is the pump laser wavelength and \( \theta \) is the half angle between the two beams. In this set-up, the angle \( \theta \) is also the same as the diffraction angle of the +1 and -1 order diffracted beams; by using the grating equation for first order diffraction - \( \lambda = 2d\sin\theta \) - the fringe spacing \( \Lambda \) can be rewritten as:

\[
\Lambda = \frac{d}{2}
\]
Figure 1.3. a) Schematic diagram of a distributed feedback dye laser (DFDL). The +1 and -1 order diffracted beams undergo total internal reflection in the quartz block and interfere with each other in the DFDL cell. This geometrical arrangement allows high visibility interference fringes to be formed even with pump lasers that have short coherence lengths. b) Schematic diagram for a DFDL capable of generating subpicosecond laser pulses.
where 1/d is the line frequency of the grating. Optical feedback or constructive interference is only satisfied for the wavelengths satisfying the Bragg scattering condition - i.e. constructive interference occurs when the scattering sites are λ/2 apart. Since the lasing wavelength λ_L is twice the fringe spacing, the wavelength of the DFDL is then given by the relation:

\[ \lambda_L = nd \]

where n is the index of refraction of the dye solution. Of course, for lasing to occur, a dye must be chosen that has gain at that wavelength. Distributed feedback dye lasers arranged in this type of set-up are capable of generating pulse widths on the order of tens of picoseconds. For subpicosecond pulse generation, a different type of DFDL must be used.

In order to produce subpicosecond DFDL pulses, the length of the interference fringe pattern in the dye cell must be less than the product of the speed of light in the dye solution and the pulse width of the laser. For typical dye solutions, this means that the interference length must be less than 200 μm. This requirement can be fulfilled with the type of DFDL shown in Figure 1.3b. After expansion by a cylindrical telescope, the pump beam passes through a coarse (55 lines/mm) transmission grating. The zeroth order transmitted beam is blocked while the +1 and -1 order diffracted beams are sent through a microscope objective to produce an interference pattern in a 1 x 10 M⁻¹ solution of Coumarin 307 in DMF. The wavelength λ_L of this DFDL is given by [2]:

\[ \lambda_L = \frac{nd}{M} \]

where M is the magnification factor of the microscope objective. By varying the distance between the microscope objective and the transmission grating, the DFDL wavelength can be tuned to twice the KrF laser wavelength. The area of the interference fringe pattern is 6 x 80 μm² [2] - an area sufficiently small for subpicosecond pulse generation. More
detailed information on the operation of DFDL's can be found in the literature [10,11,12,13].

IV. Two Stage Amplifier Section

The 500 fs pulse from the DFDL is amplified by two dye laser amplifiers separated by a saturable absorber. The first amplifier is a transversely pumped dye cell containing a 7 x 10 M⁻³ solution of Coumarin 102 in ethanol. Following the first amplifier, the DFDL pulse is focused in a Schott OG 530 glass filter which acts as a saturable absorber. The glass filter reduces the ASE content of the amplified DFDL pulse and prevents the ASE from the second amplifier from coupling to the first. The glass filter also induces a frequency chirp on the DFDL laser pulse. (The frequency chirp of the laser pulse is discussed in more detail in Chapter 2.) Using a 2 x 10 M⁻³ solution of Coumarin 307 in ethanol, the second amplifier consists of a Bethune type dye cell which results in a homogeneously pumped active medium (Figure 1.4). After the second dye amplifier, the DFDL pulse is frequency doubled in a nonlinear crystal.

V. Second harmonic generation

The second harmonic of the visible laser pulse is generated by a 200 μm thick BBO crystal. Since the second harmonic conversion efficiency is proportional to sinc(ΔkL) [14], the product of the wave vector mismatch Δk and the length L of the crystal should be made as small as possible. Because the subpicosecond pulse has a relatively wide (~5 Å) spectrum, it is difficult to satisfy phase matching conditions simultaneously for each spectral component over the entire length of the crystal. When phase matching conditions are not satisfied, the different spectral components travel with different phase velocities, leading to broadening of the laser pulse width. To solve this problem, the length of the crystal is made short enough so that pulse broadening effects are minimized. However, the
Figure 1.4. Pumping geometry for the Bethune cell amplifier. Each cylindrical lens is one focal length from the capillary tube containing the dye solution. This type of pumping geometry is used in the last dye amplifier stage in order to produce a spatially homogeneous output beam.
crystal must be made long enough so that at least 1 µJ of UV light can be generated. The BBO crystal is cut at a phase matching angle of 52° for Type I phase matching of the visible and ultraviolet laser pulses. The UV seed pulse is then sent through the KrF amplifier.

After one pass through the excimer amplifier, the UV laser pulse passes through a beam expanding telescope with a magnification of 5. A 50 µm pinhole which is contained inside an evacuated cell is placed at the focal point between the two lenses in the telescope. The pinhole cell is used as a spatial filter to reduce the ASE content in the beam and must be evacuated to prevent breakdown at the focal point of the telescope. The expanded beam leaving the telescope is then sent through the excimer amplifier again where the gain is saturated.

The final output of the laser system is a 500 fs, 10 mJ pulse centered at 248.5 nm. The UV output beam has a cross section of 10 x 30 mm² and is linearly polarized with 75% of the polarization in the vertical direction. By varying the gas pressure in the KrF excimer amplifier chamber, the timing of the gain is adjusted so that amplification of the UV seed pulse is maximized on its second pass through the amplifier (see Appendix A for details). A detailed schematic diagram of the laser system showing each of the components discussed above is found in Figure 1.5.
Figure 1.5. Detailed schematic diagram of the subpicosecond KrF excimer laser system. The dye laser system emits a 20 µJ 500 fs pulse at 496 nm. This pulse is then frequency doubled by the BBO crystal, resulting in ~2 µJ seed pulse for the excimer amplifier. The final output after amplification is a 10 mJ 500 fs pulse at 248 nm.
References


Chapter 2: Spectral Characteristics

I. Introduction

As discussed in the previous chapter, the stability of the excimer laser system is linked to the stability of the DFDL. Hence, for alignment purposes, a practical method for determining the stability of the DFDL is required. When the DFDL is pumped properly, a single, subpicosecond pulse is emitted; however, multiple pulses can be generated when the DFDL is over pumped. Fortunately, this temporal instability can be diagnosed from measurements in the spectral domain. Spectral analysis is a simple and sensitive method that can be used to align the laser system. Although temporal instability can be determined with the use of other types of instruments such as streak cameras or autocorrelators, these instruments are both more expensive and more difficult to use on a daily basis. Analysis of the UV laser spectrum indicates that the structure of the spectrum is due to self phase modulation and to the vibrational structure of the KrF upper state. To align and diagnose the laser system by spectral analysis, a one meter spectrometer with sub-angstrom resolution has been constructed.

II. Experimental Set-up

A one meter spectrometer equipped with a photodiode array detector is used for on-line observation of laser spectra. The spectrometer is constructed with a plane diffraction grating mounted in the Littrow autocollimation configuration. The electrical signal from the diode array is processed by a video circuit board and then viewed on a standard oscilloscope display.

A schematic diagram of the spectrometer is found in Figure 2.1. The spectrometer entrance slit consists of a variable width slit with a resolution of 10 μm. The photodiode
Figure 2.1. Schematic diagram for the 1 meter spectrometer. The external start signal is a 5 volt, 1 ms square pulse. The internal clock of the video board is set at 100 kHz so that one full scan of the diode array can be viewed when the oscilloscope is set at 1 ms/div.
array and its peripheral board are located directly above the entrance slit. A one meter focal length CaF2 lens is located one focal length from the entrance slit so that the diffracted light from the slit strikes the diffraction grating with parallel rays. The diffraction grating measures 34 x 34 mm², has a line frequency of 3000 lines/mm, and is operated in second order for the UV laser pulse. Because the grating is mounted in the Littrow autocollimation configuration, the diffracted light from the grating is reflected back along the incident light path. However, in order to image the diffraction pattern on the photodiode array, the grating is tilted at a slight angle with respect to the optic axis. More detailed information about the photodiode array and its use in the spectrometer can be found in Appendix B.

III. Resolution of the spectrometer

The linear dispersion and spectral resolution of the one meter spectrometer can easily be calculated [1]. The angular dispersion of the diffraction grating in the Littrow autocollimation mount is given by:

\[ \frac{d\theta}{d\lambda} = \frac{2\tan\theta}{\lambda} \]

where \( \lambda \) is the wavelength of the incident light and \( \theta \) is the angle angle between the normal of the grating and the optic axis. For autocollimation, the grating equation is given by \( 2d\sin\theta = n\lambda \), where \( d \) is the spacing between grating lines and \( n \) is the diffraction order. By combining the two equations, the angular dispersion can be written as:

\[ \frac{d\theta}{d\lambda} = \frac{n}{d\cos\theta} \]

The linear dispersion of the spectrometer is given by the product of the angular dispersion and the effective focal length \( f \) of the system:

\[ \frac{dx}{d\lambda} = \frac{nf}{d\cos\theta} \]
In the plane of the diode array, the dispersion per individual diode is the product of the reciprocal linear dispersion and the diode spacing:

$$\frac{d\lambda}{d(\text{diode})} = \frac{d\lambda}{25\mu m} \times \frac{25\mu m}{dx \text{ diode}}$$

Using a one meter focal length lens to image the second order diffraction of 248.5 nm light diffracted from a grating having 3000 lines/mm, the reciprocal linear dispersion of the spectrometer is 0.03 Å/diode. Since there are 1024 diodes in the array, a 28 Å wide spectrum centered at 2485 Å can be viewed with the diode array.

The spectral resolution of the spectrometer is a function of the width \( b \) of the entrance slit, the diffraction order \( m \) of the grating, and the number \( N \) of grating lines illuminated. Since the resolving power \( R \) of a grating is given by \( R = \frac{\lambda}{\Delta\lambda} = mN \), and since the number of lines illuminated on the grating is \( N = 2fsin(\text{arcsin}(\lambda/b)) \), the resolution of the spectrometer is:

$$\Delta\lambda = \frac{\lambda}{2mf \tan(\text{arcsin}(\lambda/b))}$$

For the parameters given above in the calculation of the dispersion, the resolution of the spectrometer expressed in angstroms per slit width is 0.8 Å/mm. For example, a 100 µm slit width gives a resolution of 0.08 Å. Spectra obtained from a conventional nanosecond KrF excimer laser with and without injection locking are shown in Figure 2.2. The line width of the laser with injection is believed to be limited by the resolution of the spectrometer.

IV. Experimental Results

The spectral behavior of the subpicosecond KrF excimer laser system is determined with the use of the one meter spectrometer. For the spectra examined below, the resolution of the spectrometer is 0.3 Å and the absolute wavelength scale has been calibrated against the ASE spectrum of the KrF excimer amplifier which is known to have a peak at 2485 Å.
Figure 2.2. Spectra obtained from a conventional nanosecond KrF excimer laser with a) and without b) injection locking. Part b shows the output spectrum of the amplifier and a) the output of the amplifier when a narrow line width seed beam is injected. The width of the line in a) is limited by the resolution of the spectrometer.
Under normal operating conditions, the spectrum of the amplified DFDL pulse and the twice amplified excimer laser pulse are as shown in Figures 2.3 and 2.4. Figure 2.3 shows the output of the Bethune cell amplifier when the DFDL is pumped at 2 times its threshold for lasing. The amplified DFDL spectrum has a full width at half maximum (FWHM) of 7 Å and an asymmetric shape with a tail toward the shorter wavelengths. Spectra of the subpicosecond KrF excimer laser and of the ASE from the excimer amplifier are shown in Figure 2.4. The ASE has a smooth, symmetric shape and is centered at 2485 Å. In contrast, the spectrum of the UV laser pulse has a double peaked structure and a FWHM of 6 - 7 Å. The laser spectrum also shows a long tail toward the longer wavelengths and a dip located at twice the frequency of the DFDL. The shape of the UV laser spectrum depends on the pump level of the DFDL, the intensity of the amplified pulse, and the tuning of the DFDL.

The spectrum of the amplified DFDL changes dramatically when the DFDL is pumped more than two times threshold. When the DFDL is pumped at least three times threshold, the amplified DFDL spectrum becomes modulated and varies from shot to shot. An example of such a spectrum is located in Figure 2.5a. The narrow modulation features superimposed on the normally smooth spectrum have a width of 0.6 - 0.8 Å. Additionally, there is a small feature near 4985 Å that is separated from the center spectral feature. Since the spectrum in Figure 2.5a is of the amplified DFDL, the spectral modulation of the unamplified DFDL pulse may be slightly different. When the DFDL is over pumped, the dip in the UV laser spectrum becomes slightly deeper than when the DFDL is pumped 2 times threshold and the UV laser spectrum begins to fluctuate. An example of the type of spectral fluctuation is shown in Figure 2.5b.

The spectral shape of the amplified UV laser pulse is also dependent on the intensity of the laser. In Figure 2.6, a series of spectra associated with varying pump levels of the
Figure 2.3. Spectrum of the amplified DFDL laser pulse.
Figure 2.4. Spectra of the UV laser pulse a) and the ASE from the KrF excimer amplifier b). Part a) is a characteristic laser pulse spectrum when the DFDL is pumped 2 times threshold.
Figure 2.5 Part a) and b) shown laser spectra when the laser system is unstable. Multiple pulse generation by the DFDL produces the modulated spectrum in a). The UV laser spectrum associated with the unstable DFDL is shown in b). By looking for these characteristic spectra, the laser system can be fine tuned for stable operation.
**Figure 2.6.** UV laser spectra as a function of the Bethune cell pumping. Spectrum a) shows the normal UV laser spectrum. As the pumping of the Bethune cell is reduced from b) to e), the dip in the laser spectrum becomes less pronounced. Notice that spectrum e) has a peak located at the valley in spectrum a). Spectrum f) is of the ASE from the amplifier.
Bethune cell amplifier is shown under the condition that the DFDL is pumped two times threshold. As the pump length of the Bethune cell amplifier decreases, the stronger of the two peaks in the laser spectrum shifts to longer wavelengths and the dip in the spectrum becomes less pronounced until it eventually disappears. Spectra a-c in Figure 2.6 are indicative of saturated amplification in KrF excimer amplifier. When the energy of the UV laser pulse is below saturation, the spectral shape becomes more symmetric and the peak wavelength shifts to the short wavelength side of the peak ASE wavelength. The low intensity spectra d-e have a peak near 2484 Å. This indicates that the DFDL wavelength associated with all of the spectra in Figure 2.6 is twice the wavelength of 2484 Å, or 4968 Å. Spectrum e is a reference spectrum of the KrF amplifier ASE.

Variation in the UV laser spectrum as a function of the tuning of the DFDL, and, thus, as a function of the wavelength of the UV seed pulse, is shown in Figures 2.7 and 2.8. For the spectra in these figures, the DFDL is pumped 1.5 - 2 times threshold and the BBO crystal has been optimized for producing the normal, double peaked UV laser spectrum. The normal spectral shape of the UV laser pulse is shown in Figure 2.7a. When the UV seed pulse is tuned a few angstroms to the short or long wavelength side of 2484 Å, the two peaks in the laser spectrum become nearly equal in intensity. Figure 2.8 shows the effect on the UV laser spectrum when the UV seed pulse is tuned to the long wavelength wing of the gain. On the long wavelength side of the gain bandwidth, the laser spectrum becomes singly peaked and peak intensities are observed when the laser is tuned to 2488 Å or 2492 Å. When the laser is tuned between these two wavelengths, the spectrum has two relatively weak peaks near 2488 Å and 2492 Å that are comparable in intensity to the ASE background. Note that the intensity scales for the spectra in Figures 2.7 and 2.8 are directly comparable and that the location and relative intensity of the ASE is included for reference.
Figure 2.7. Variation of the UV laser spectrum near the center of the KrF gain. The normal asymmetric spectral shape of the laser spectrum is shown in a). The ASE from the excimer amplifier is shown in b).
Figure 2.7.(continued) Variation of the UV laser spectrum near the center of the KrF gain. When the laser is tuned a few angstroms to shorter wavelengths c) or longer wavelengths d) the two peaks becomes nearly equal in intensity.
Figure 2.8. Variation of the UV laser spectrum near the long wavelength edge of the gain. As the laser is tuned away from the center of the gain, the laser spectrum a) becomes singly peaked. Spectrum b) shows two low intensity peaks comparable to the background ASE.
Figure 2.8. (continued) Variation of the UV laser spectrum near the long wavelength edge of the gain. As the laser is tuned to even longer wavelengths, a peak c) in the output of the laser is observed when the laser is tuned to 2492 Å. Part d) shows the ASE spectrum for reference.
V. Discussion

The spectral characteristics of the KrF excimer laser system are the result of a combination of factors: the pump level of the DFDL, spectral broadening by the OG 530 saturable absorber, second harmonic conversion by the BBO crystal, and saturated amplification of the UV laser pulse in the excimer amplifier. Although it is unknown precisely how each of these factors influences the spectral characteristics of the system, observation of the laser spectra does provide a simple method for fine tuning and stabilizing the laser system. In addition to the factors mentioned above, self-phase modulation and the electronic structure of the KrF excimer molecule are shown to influence the spectral shape of the final output pulse. Near the center of the excimer gain bandwidth, self phase modulation causes spectral modulation. When the UV seed pulse is tuned near the edge of the gain bandwidth, the laser spectrum is influenced by the vibrational structure of the KrF upper state.

As shown in Figure 2.5a, the DFDL spectrum becomes modulated when the pump level of the DFDL is too strong. One consequence of this observation is that the output of the DFDL can be stabilized by simultaneously observing the DFDL spectrum and changing the DFDL pump energy until no spectral modulation is present. [The pump level of the DFDL can be adjusted by changing the pumping of the final amplifier in the picosecond dye laser section. A detailed description of the adjustment process is found in Appendix A.] The observed spectral modulation is a signature of multiple pulse generation by the DFDL. When the DFDL emits multiple pulses, the individual pulses interfere with one another and produce a modulated spectrum where the modulation period (Δλ) decreases with increasing separation (ΔT) between the pulses. This relationship can be analytically modeled by the function \( \Delta \lambda = \frac{\lambda^2}{(c\Delta T)} \) [2]. Figure 2.9 describes how this formula can be derived from the Fourier transform of a periodic signal. For example, the 1.4 Å modulation period for the spectrum in Figure 2.5a corresponds to a separation of 5.9 ps
\[ x(t) = \sum_{k=-\infty}^{\infty} \delta(t - kT) \]

Fourier transform pair

\[ X(\omega) = F(x(t)) = \frac{2\pi}{T} \sum_{k=-\infty}^{\infty} \delta\left(\omega - \frac{2\pi k}{T}\right) \]

Uncertainty relation

\[ \Delta\omega \Delta T = 2\pi \]

or

\[ \Delta\lambda \Delta T = \frac{\lambda^2}{c} \]

Figure 2.9. The Fourier transform of a periodic delta function signal (A) in the time domain leads to a periodic delta function signal (B) in the frequency domain. An expression for the uncertainty principle gives the function for modelling the spectral structure in the DFDL spectrum.
between the main DFDL pulse and the trailing pulse. Even when the spectrum of the DFDL is modulated, the spectrum of the second harmonic pulse shows no modulation. However, the spectrum of the amplified UV laser pulse becomes unstable and varies from shot to shot. The asymmetry and width of the normal DFDL spectrum (see Figure 2.3) is a sign of frequency chirping, the validity of which will be discussed in the next section.

The most striking feature of the UV laser spectrum is its double peaked structure. The valley between the two peaks is located at twice the frequency of the DFDL center frequency, and, from the series of spectra in Figure 2.6, the valley is shown to become more pronounced as the energy of the laser pulse increases. Furthermore, the double peaked structure is not observed when the OG 530 saturable absorber is removed from the laser system. It is possible that non-saturable absorbers such as Kr$^+$, Ar$^+$, F$^-$, Kr** and Ar** [3] could play a role in defining the structure of the UV laser spectrum. However, the double peaked structure varies smoothly and continuously as the UV seed pulse is tuned a few angstroms to either side of 2484 Å. If the double peaked structure is due to non-saturable absorption in the KrF gain medium, then the location of the absorption feature - the valley - should remain constant as the laser wavelength is tuned; but this is not observed. The width of the laser spectrum is roughly twice that of the ASE spectrum from the KrF amplifier. This broadening is at least partially due to the broadening that is caused by saturated amplification in an inhomogeneously broadened gain medium.

From the preceding observations, it can be postulated that some non-linear amplification process is responsible for the double peaked structure in the laser spectrum. The process of self-phase modulation (SPM) is a likely candidate for describing the observed spectral behavior since SPM causes modulated spectra in which the degree of modulation varies directly with the intensity of the laser pulse. Additionally, SPM leads to frequency chirping and, as mentioned in the Introduction, the output of the KrF excimer
laser system is known to be frequency chirped. SPM is the result of an intensity dependent refractive index that is created when an intense laser pulse travels through almost any medium. The changing refractive index modifies the phase of the laser pulse, leading to time dependent frequency broadening.

The process of SPM can be explained by the following model [4]. Let the electric field of the laser pulse be represented by \( E = E(z,t)\exp(\,i\,\phi(t)\,) \), where the phase factor is given by:

\[
\phi(t) = \omega_L t - k_L z = \omega_L - \frac{\omega_L n(t)}{c}
\]

If an intensity dependent refractive index is defined as \( n(t) = n_0 + n_2 |E|^2 \), then the phase of the laser pulse is given by:

\[
\phi(t) = \omega_L t - \frac{\omega_L n_0}{c} - \frac{\omega_L n_2 |E|^2}{c}
\]

Lastly, the instantaneous frequency is defined as the time derivative of the phase so that:

\[
\omega = \frac{d\phi}{dt} = \omega_L - \frac{\omega_L^2}{c} \frac{d}{dt}[n_2 |E|^2]
\]

\[
\Delta \omega = \omega - \omega_L = - \frac{\omega_L^2}{c} \frac{d}{dt}[n_2 |E|^2]
\]

where \( \Delta \omega \) denotes the spectral broadening. A graphic representation of this model is shown in Figure 2.10. From the model just described, it is apparent that SPM leads to spectral broadening where the degree of broadening varies inversely with the pulse width of the laser. SPM also leads to spectral structure that reflects the temporal symmetry or asymmetry of the laser pulse. As shown in Figure 2.10, the frequency broadening may not be a single valued function of time. Consequently, each frequency may occur twice and it is the interference between the two occurrences of a given frequency that gives rise to spectral modulation.

The double peaked structure of the UV laser pulse can be modelled using self phase modulation. Let the electric field of the laser pulse be described by the equation:
Figure 2.10 Schematic diagram of SPM with a symmetric, asymmetric, and short pulse envelope. Row A graphs the laser pulse envelopes, row B the time derivative of the envelopes, and C the spectral broadening. It is apparent from the model that the degree of spectral broadening is inversely proportional to the pulse width and that the spectral shape is influenced by the symmetry or asymmetry of the pulse envelope.
\[ E(t) = E_0(t)e^{\{-i(\omega_0 t + \phi(t))\}} \]

where \( E_0(t) \) is the envelope of the pulse shape, and \( \phi(t) \) is the nonlinear phase produced by the intensity dependent refractive index. The power spectral density \( S(\omega) \) of the phase modulated pulse is then proportional to the Fourier transform of the electric field \( E(\omega) \) [5]:

\[ S(\omega) = \frac{\varepsilon_0 c}{2} |E(\omega)|^2 \]

\[ E(\omega) = \mathcal{F}(E(t)) = \int \exp\{i\omega t\} E_0(t)e^{\{-i(\omega_0 t + \phi(t))\}} dt \]

If the pulse envelope is defined as the Gaussian function \( E_0(t) = \exp\{- (t/T)^2 \} \) and the nonlinear phase is defined as \( \phi(t) = \beta|E_0(t)|^2 \), then the power spectrum will be symmetric and the degree of modulation will be influenced by the pulse width \( T \) and the the nonlinear coefficient \( \beta \). Figure 2.11 shows a series of spectra for a \( T = 500 \) fs pulse with varying nonlinear phase coefficients. As \( \beta \) increases, the degree of SPM increases and the spectrum becomes multi-peaked. Notice also that the valley between the two peaks is located at the peak wavelength of the low \( \beta \) spectra. This type of behavior is observed in the spectrum of the UV laser pulse as the pumping of the Bethune cell amplifier is changed. The main difference between the calculated and observed spectra lies in the asymmetry of the observed spectra.

In order to reproduce the asymmetry in the observed spectra, an asymmetric pulse envelope can be used in the SPM model. By defining the pulse envelope as \( E_0(t) = (t/T)e^{\{- (t/T)^2 \}} \) and the nonlinear phase coefficient as \( \phi(t) = \beta|E_0(t)|^2 \), the calculated spectra become similar to the experimentally observed spectra as can be seen by comparing the spectra in Figures 2.6 and 2.12. As the nonlinear phase coefficient \( \beta \) increases, the strong peak in the calculated spectrum moves toward shorter wavelengths, and the valley between the two peaks shifts to the peak wavelength of the the low \( \beta \) spectra. Recall, that this is the same type of behavior that is experimentally observed for the spectra shown in Figure 2.6. Thus, by changing the pump level of the Bethune cell amplifier, the degree of
Figure 2.11 Power spectrum of self phase modulated symmetric Gaussian pulse envelopes. Spectrum a) corresponds to $\beta=0$. Spectra b) and c) correspond to $\beta$ values of 6 and 8, respectively, or phase shifts of $1.9\pi$ and $2.6\pi$ during a pulse width $T=500$ fs. The spectra are computed using the FFT function of a computer software program called Matlab.
Figure 2.12 Power spectrum of self phase modulated asymmetric Gaussian pulse envelopes. Spectrum a) corresponds to \( \beta = 0 \). Spectra b) and c) are for \( \beta \) values of 15 and 25, respectively, or phase shifts of \( 4.8\pi \) and \( 8\pi \) during a pulse width \( T = 500 \) fs. The spectra are computed using the FFT function of a computer software program called Matlab.
SPM in the UV laser pulse can be controlled.

Although self-phase modulation can account for the shape of the UV laser spectrum when the laser is tuned near the center of the KrF gain curve, the model of the laser spectrum needs to be modified when the laser is tuned to the wings of the KrF gain curve. As indicated in Figure 2.8, the spectrum of the UV laser pulse becomes singly peaked when the laser is tuned near 2490 Å. Note that the broad feature in the spectra near 2485 Å is due to ASE from the amplifier and not to the laser. Maxima of the laser spectrum are observed when the laser is tuned to either 2488 Å or 2492 Å. This behavior is indicative of resonance with individual excited state vibrational levels in the upper state of the lasing transition. The transition in KrF near 248 nm occurs between a bound upper state and a repulsive lower state [6]. The potential energy diagram in Figure 2.13 shows the relevant energy levels for the KrF(B→X) transition. The contribution of each vibrational state in the upper state to the overall fluorescence spectrum of KrF can be calculated according to the reflection method [7]. A general representation of the reflection method is presented in Figure 2.14. By approximating the wavefunctions of the upper vibrational states by simple harmonic oscillator wavefunctions, the emission line shape from a given vibrational level is obtained by reflecting the probability density of the upper state wavefunction at the lower state potential curve. The reflection method can be used to calculate the emission spectrum of the KrF(B→X) transition since the change in the lower state potential in the vicinity of the equilibrium internuclear distance is larger than the vibrational energy $\hbar \omega$ of the upper state.

The lower state potential energy curve can be described by a Born–Mayer potential $V_L$:

$$V_L = e_0 \exp \left( -\alpha (R - R_0) \right)$$

where $R$ is the internuclear distance between atoms, and $e_0$ is the energy of the lower state potential at the equilibrium internuclear distance $R_E$. Using harmonic oscillator
Figure 2.13 Potential energy diagram for the B to X transition in KrF. The upper state vibrational energy is 310 cm⁻¹ and the equilibrium internuclear distance of the upper state potential well is 2.27 Å. When the upper state decays to the lower repulsive state, a photon is given off near 248 nm.
Figure 2.14 Diagram of the reflection method for calculating the line shape $g(E)$ due to the first vibrational level of the upper electronic state of an excimer transition. In the vicinity of $R_e$, the upper potential $V_U(R)$ is approximated by a harmonic oscillator potential. The line shape is calculated by reflecting the harmonic oscillator wavefunctions at the lower state potential $V_L(R)$. 

wavefunctions for the upper state, the line shape of each vibrational energy level can be calculated by the following formula [7]:

\[
g_{v}(E) = \frac{k}{\alpha E} \frac{1}{\sqrt{v!}} \sqrt{\frac{\pi}{2}} H_{v}^2 \left( -\frac{k}{\alpha \varepsilon_0} E \right) \exp \left\{ -\left( \frac{k}{\alpha \varepsilon_0} E \right)^2 \right\}
\]

where \( k = (\mu \omega / h)^{1/2} \) and \( H_{v} \) is the Hermite polynomial of degree \( v \). The characteristic vibrational frequency of the upper state is \( \omega \) and \( \mu \) is the reduced mass of KrF.

Using the values of \( \omega = 310 \text{ cm}^{-1} \) and \( R_e = 2.27 \text{ Å} \) that were obtained by Tellinghuisen et al [8], the contribution of the \( \nu = 0, 1, \) and \( 2 \) vibrational states to the overall emission spectrum can be calculated. Figure 2.15 shows the result of such a calculation with \( \alpha = 2.02 \text{ Å}^{-1} \) and \( \varepsilon_0 = 750 \text{ cm}^{-1} \). In order to calculate the overall fluorescence line shape, the product of the individual vibrational state line shapes and their relative populations must be added. Since \( \alpha \) determines the slope of the lower potential, the calculation depends sensitively on the value of \( \alpha \). Near the wavelength of the KrF B->X transition, the calculation shows that the contribution of the \( \nu = 1 \) and \( \nu = 2 \) vibrational states peaks at 2488 Å and 2492 Å, respectively. The experimentally observed behavior in the spectrum can be explained by postulating that the laser intensity increases when the laser is tuned to the peak, or resonance, wavelength associated with either the \( \nu = 1 \) or \( \nu = 2 \) vibrational state. The spectral behavior of a subpicosecond XeCl excimer laser has been observed to depend on vibrational energy level splitting [9,10]. However, for XeCl, the spectral structure is due to the small vibrational frequency (26 cm\(^{-1}\)) of the weakly bound lower state and not to the vibrational structure of the upper state.

Spectral characteristics due to the vibrational structure of KrF near 2491 Å have also been observed in conventional, nanosecond KrF excimer lasers. KrF laser spectra with two distinct peaks - near 2484 Å (strong peak) and 2491 Å (weak peak) - have been observed for both e-beam [11] and discharge pumping sources [12]. The two peaks in
Figure 2.15 Contribution of the $v=0$, 1, and 2 upper state vibrational levels to the KrF($B\rightarrow X$) transition. On the long wavelength side of the gain bandwidth, the laser intensity increases when the laser is tuned to resonance with the $v=1$ or $v=2$ vibrational state. When the laser is tuned between the peak contributions of the $v=1$ or $v=2$ state, the laser spectrum appears as shown in Figure 2.8b.
such spectra are believed to be caused by separate lasing from the \( v = 0 \) and \( v = 1 \) vibrational states. Alternatively, some have proposed that the dip in the fluorescence and laser spectra is due to transient absorption.
References


Chapter 3: Single Shot Ultraviolet Autocorrelator

I. Introduction

Since electronic detection techniques with femtosecond resolution are unavailable, autocorrelation techniques are often used to measure the pulse width of ultrashort pulse lasers. The basic principle of an autocorrelation technique is to convert information about the temporal duration of the laser pulse into spatial information that can be detected by a spatial detector (see Figure 3.1). Ultrafast laser pulse width measurements have traditionally been measured using multiple shot autocorrelation techniques in non-linear crystals. For an autocorrelator set up in the Michelson interferometer configuration, the autocorrelation trace of the laser pulse is built up by recording the second harmonic intensity of the light generated by the non-linear crystal while varying the overlap between the two pulses emerging from each arm of the interferometer. By varying the length of one arm of the interferometer, the two replicas of the incident laser pulse can be overlapped and the time axis of the autocorrelation can be calibrated.

Single shot autocorrelators have also been developed so that temporal measurements can be obtained from a single laser pulse. Such methods are especially valuable for ultrashort pulse laser systems operating at low repetition rates. Single shot autocorrelators used in the visible part of the spectrum that are based on second harmonic generation in nonlinear crystals or two photon fluorescence in dyes [1,2] are not directly transferable to ultraviolet laser pulses. Techniques based on second harmonic generation in nonlinear crystals may not be used since available nonlinear crystals are neither phase matchable for second harmonic generation nor transparent in the vacuum ultraviolet. Figure 3.2 shows the spectral range and short wavelength limit for phase matchable second harmonic generation for several nonlinear crystals [3]. From the chart it is apparent that the lowest fundamental wavelength for which second harmonic generation occurs is at 410 nm for
Figure 3.1 Schematic diagram of a multiple shot autocorrelator set up in a Michelson interferometer configuration. By changing the length of one arm of the interferometer, the overlap of the two output pulses can be varied. The second harmonic light generated in the nonlinear crystal is proportional to the degree of overlap of the two output pulses.
Figure 3.2. This chart of nonlinear crystals shows the spectral range of fundamental wavelengths that are phase matchable for second harmonic generation. The shortest ultraviolet wavelength (∼200 nm) is reached by second harmonic generation in a BBO crystal [3].
BBO crystals with Type I phase matching. Because dye molecules have absorption bands in the ultraviolet, two photon absorption of visible laser pulses and the resulting visible dye fluorescence can be used for autocorrelation of visible laser pulses. However, this nonlinear process is not applicable for ultraviolet lasers since the dye already absorbs at the wavelength of the laser.

Autocorrelation methods for ultraviolet lasers are usually based on multiphoton ionization or fluorescence from decaying diatomic molecules. A list of nonlinear media that have been used for autocorrelation of ultraviolet laser includes: three photon fluorescence of XeF [4,5], two photon fluorescence of Xe2 [6], two photon fluorescence of Cd vapor [7], two photon fluorescence from self trapped excitons in CaF2 [8], two photon luminescence of CsI doped with Na [9], two photon ionization of NO [10], two photon ionization of gases [11], and two photon absorption in diamond [12].

II. Method

A single shot autocorrelator based on the fluorescence of XeF was developed to measure the pulse width of the subpicosecond KrF excimer laser [4]. A schematic diagram of the autocorrelator is found in Figure 3.3 A beamsplitter produces two copies of the laser pulse that are crossed at a small angle in a pressure cell containing Xe, F2, and Ar. The cylindrically focused subpicosecond excimer laser pulse induces both XeF(B->X) (351 nm) and XeF(C->A) (480 nm) fluorescence in the gas mixture [5]. The XeF excimer is produced by three photon ionization of Xe and the subsequent reaction of Xe+ with F-. When the two pulses overlap spatially and temporally, the spatial distribution of the XeF fluorescence yields the autocorrelation function of the incident laser pulse. The visible fluorescence pattern of the XeF C->A transition is recorded by a CCD camera.

The spatial distribution of the XeF fluorescence pattern produces a third order autocorrelation function along the vertical axis formed by the overlap of the two beams.
Figure 3.3 Schematic diagram of a single shot autocorrelator for the subpicosecond KrF excimer laser. The spatial distribution of the XeF fluorescence is proportional to a third order autocorrelation function.
The autocorrelation function is third order because of the cubic dependence of the XeF fluorescence intensity on the KrF laser intensity [5]. Since the ionization potential of Xe is 12 eV, three 5 eV photons from the subpicosecond KrF excimer laser have enough energy to form Xe$^+$ when the intensity of the laser is high enough for multiphoton ionization:

$$\text{Xe} + 3(h\nu=5 \text{ eV}) \rightarrow \text{Xe}^+ + e^-.$$  

The XeF(B) state is formed by the reaction of a xenon ion and a fluorine ion created by electron attachment of F$_2$:

$$\text{Xe}^+ + \text{F}^- + \text{Ar} \rightarrow \text{XeF(B)} + \text{Ar}.$$  

The XeF(C) state can be populated by collisions between Ar and the XeF(B) state excimer [13]:

$$\text{XeF(B)} + \text{Ar} \leftrightarrow \text{XeF(C)} + \text{Ar}.$$  

Visible and ultraviolet fluorescence occur when the XeF(B) and XeF(C) states decay:

$$\text{XeF(B)} \rightarrow \text{Xe} + \text{F} + h\nu(351 \text{ nm})$$

$$\text{XeF(C)} \rightarrow \text{Xe} + \text{F} + h\nu(480 \text{ nm}).$$  

The time duration of the fluorescence induced by the subpicosecond KrF excimer laser has been measured to be a few nanoseconds [8]. The fluorescence time is short enough so that spatial diffusion of the XeF excimer does not effect the resolution of the autocorrelation technique.

The time averaged autocorrelation function $a_2(\tau)$ that is recorded is proportional to:

$$a_2(\tau) = \left\langle \left( \int |E(t)|^2 \, dt \right)^2 \right\rangle$$

where $E(t) = E_1(t) + E_2(t-\tau)$ is the sum of the electric fields of the two pulses entering the gas cell and $\langle \rangle$ denotes time averaging. In order to relate the spatial distribution of the XeF fluorescence pattern to the temporal width of the autocorrelation function, a delay line is inserted in one of the beam paths. By introducing a delay $\Delta T$ in the path of one of the
beams, the maximum of the XeF fluorescence pattern shifts by $\Delta X$ (see Figure 3.4). The shift $\Delta X$ is related to the delay $\Delta T$ by the expression [4]:

$$\Delta X = \frac{c\Delta T}{2n\sin(\phi/2)}$$

where $n$ is the index of refraction of the gas and $\phi$ is the angle between the two beams. For example, if the angle between the two beams is 3°, $\Delta T$=500 fs, and $n=1$, then the maximum of the XeF pattern shifts by 3 mm. The full width at half maximum (FWHM) $S$ of the XeF spatial distribution is related to the FWHM $A$ of the autocorrelation function by $A = S\Delta T/\Delta X$. The width of the autocorrelation function can then be used to determine the pulse width $P$ of the incident pulse by applying the relation $P = KA$, where $K$ is the deconvolution factor that depends on the incident pulse shape. In general, $K$ is the ratio of the width of the assumed pulse shape to the width of the autocorrelation function of the assumed pulse shape. For example, $K = 1$ if a square pulse shape is assumed, or $K = .775$ if the pulse shape is described by a sech$^2(t)$ function and the autocorrelation method is third order.

III. Experimental arrangement

The physical layout of the single shot autocorrelator is shown in Figures 3.3 and 3.5. Figure 3.5 shows only the lower beam path in Figure 3.3. A 20 x 10 mm$^2$ area of the KrF beam with about 2 mJ of energy is selected as the input beam to the autocorrelator. An area smaller than the total area of the KrF beam is selected to ensure that the input pulse is spatially uniform. The input pulse enters the autocorrelator and is split by a 50% beamsplitter. The transmitted beam passes through a variable delay line and is directed toward the XeF cell. The reflected beam enters the XeF cell at an angle of 3° with respect
Figure 3.4. This diagram shows the geometrical arrangement of the beams inside the XeF cell. In a), the XeF fluorescence occurs where beam 1 and 2 overlap spatially and temporally. In b), beam 2 is delayed with respect to beam 1, so the fluorescence pattern shifts by $\Delta X$. c) shows that the shift $\Delta X$ is related to the delay $\Delta T$ by $\Delta X = c\Delta T/2\sin(\phi/2)$. 
Figure 3.5 Physical arrangement of the autocorrelator showing only the beam path that is transmitted through the beamsplitter. See Figure 3.3 for a three dimensional view.
to the axis of the transmitted beam. A microposition stage with a resolution of 2 \( \mu \text{m} \) is used to introduce a known path length difference between the transmitted and reflected beam. Both transmitted and reflected beams are focused by a cylindrical lens \( (f = 15 \text{ cm}) \) to the center of the XeF cell. The XeF cell itself is made of 1.5" diameter stainless steel pipe and four CaF2 windows which were chosen because of their compatibility with fluorine environments. The gas inside the cell consists of 1.00 atm of Xe, 0.425 atm of 10 \% F2 in Ar, and 2.15 atm of Ar. The XeF fluorescence pattern is observed perpendicular to the propagation direction of the KrF beams.

The detection system consists of imaging optics, a CCD camera, and graphics processing software. In order to image the XeF fluorescence pattern on the CCD camera, two reverse mounted 35 mm camera lenses are used. This arrangement allows the maximum amount of light to be transmitted from the object plane to the image plane and reduces spherical aberration effects. The Cohu CCD camera and camera lenses act as a UV cut off filter and, thus, permit only the XeF(C->A) fluorescence to be observed. Once the XeF fluorescence pattern is captured by the video system, graphics processing software (Beamcode) is used to obtain the autocorrelation trace.

IV. Results

A typical autocorrelation trace obtained from this autocorrelator is shown in Figure 3.6. The autocorrelation trace is calibrated by observing the number of pixels that the peak of the XeF fluorescence shifts across the video screen for a given delay introduced with a translational stage. For example, if the delay line is moved 75 \( \mu \text{m} \), then a 150 \( \mu \text{m} \) path length difference or a 500 fs delay between the two beams is introduced. By noting the change in pixel location of the fluorescence pattern peak intensity, the calibration factor \( \Delta T/\Delta X \) can be obtained. With the present imaging system, the calibration factor is approximately 7 fs/pixel with an estimated error of 10\%. A false color image of the XeF
Figure 3.6: Single shot intensity autocorrelation trace. The third order autocorrelation function has a width of 616 fs. Assuming a sech$^2(t)$ shaped pulse, the pulse width is 477 fs. The peak to background intensity ratio is nearly 10 to 1.
fluorescence pattern detected by the CCD camera is shown in Figure 3.7. Using the Beamcode graphics software, the autocorrelation trace is obtained by making a profile of the image along the axis of greatest intensity. The FWHM of the autocorrelation trace obtained in this manner is 616 fs. Assuming a third order autocorrelation function based on a sech²(t) pulse shape, the FWHM of the KrF laser pulse is 477 fs. The peak to background ratio of the autocorrelation trace is in agreement with the theoretically predicted value of 10 to 1 for third order autocorrelation techniques [14].

The quality of the observed autocorrelation trace depends on a number of factors. In 500 fs, the laser pulse only travels a distance of 150 μm. Therefore, temporal and spatial overlap of the two beams entering the XeF cell is very sensitive to the length of the delay line and the alignment of the beams. When the two beams are overlapped in time and space, the XeF fluorescence can easily be seen by dark-adapted eyes. Fine tuning of the overlap between the two beams can be adjusted by placing a thin quartz plate in one of the beam paths. Because the autocorrelation technique is based on three photon ionization of Xe, the XeF fluorescence intensity is a sensitive function (cubic) of the intensity of the two beams entering the XeF cell. The energy in each beam must be at least 1 mJ in order for the CCD camera to detect any XeF fluorescence. Consequently, after mirror losses are considered, most of the output energy from the laser must be used to make an autocorrelation trace. This problem limits experiments that are based on simultaneous measurement of the pulse width and phenomena dependent on high intensities. The best autocorrelation traces are obtained after a new gas mixture is made for the KrF excimer amplifier. The gas inside the XeF cell shows little degradation after sitting six months in a well passivated cell.

Because second order intensity autocorrelation functions are inherently symmetric functions, the symmetry or asymmetry of the incident laser pulse can not be determined. To determine the symmetry, higher order autocorrelations must be performed in addition to
Figure 3.7. XeF fluorescence pattern. This figure shows a false color image of the XeF fluorescence detected by the CCD camera. The autocorrelation trace in Figure 3.6 is obtained by making a horizontal profile along the axis of greatest intensity.
the second order autocorrelation. Etchepare et al [15] report that third order autocorrelation measurements can reveal information about the asymmetry of the original pulse. Careful examination of the autocorrelation trace in Figure 3.6 shows that the function is slightly asymmetric with respect to the location of the peak intensity. The asymmetry in the autocorrelation function implies that the original pulse is also asymmetric. This observation supports the postulate presented in Chapter 2 to explain the asymmetric shape of the laser spectrum. However, it is possible that the measured asymmetry in the autocorrelation trace could be due to some spatial non-uniformity of the original laser pulse.
References


Chapter 4: Single Shot Phase Sensitive Autocorrelator

I. **Introduction**

In addition to spectral and temporal measurements, complete characterization of an ultrafast pulse laser system includes determining the phase of the laser pulse. This information is of importance because it is by manipulating the phase of the laser pulse that the shortest subpicosecond laser pulses are generated. The shortest pulses are generally produced by balancing two opposing effects: nonlinear frequency chirping caused by self phase modulation (SPM) and negative group velocity dispersion caused by a dispersive delay line. For example, the former effect can be accomplished by propagating a laser pulse through an optical fiber, while the latter accomplished by a grating or prism pair. In order to design the appropriate delay line to match the frequency chirp caused by SPM, the functional form of the time varying phase must be known. Unfortunately, though, traditional intensity autocorrelation techniques are unable to determine the phase of the laser pulse.

Many different types of chirp sensitive measurements have been developed for ultrafast visible laser pulses [1-8]. When traditional second order intensity autocorrelation techniques are performed with interferometric accuracy, the chirp of the laser pulse can be indirectly inferred from the autocorrelation trace by iteratively fitting an assumed pulse shape to the observed autocorrelation trace [4,8]. Another, more direct method, is to perform a cross correlation between a transform limited short pulse and a frequency chirped pulse [1,2,6]. Each of these techniques, however, relies on the use of nonlinear crystals that can not be used for autocorrelation of ultraviolet laser pulses. The frequency sweep of an ultraviolet continuum generated by focusing a subpicosecond KrF laser pulse in a high pressure gas has been determined by Glownia et al [9]. In the experiment, the frequency sweep is not determined by an autocorrelation measurement, but is measured by
a multiple shot pump-probe experiment based on photodissociation of TiCl molecules. The goal of this chapter is to present the design of a single shot phase sensitive autocorrelator for ultrafast ultraviolet laser pulses. The design of the autocorrelator involves combining the autocorrelation methods developed for ultraviolet lasers with the chirp sensitive autocorrelation techniques designed for visible lasers.

The results of the spectral and pulse width measurements of the subpicosecond KrF excimer laser suggest that the laser pulse is frequency chirped. Given a spectral width of 5 Å and a pulse width of 477 fs, the time-bandwidth product - \( \Delta \nu \Delta \tau \) - of the UV laser pulse is 1.2. Since the time-bandwidth product for a transform limited sech\(^2\)(t) pulse is 0.315 [10], the bandwidth of the subpicosecond KrF laser will support a pulse shorter than 477 fs. The modulation and asymmetry of the laser spectrum also support the conclusion that the laser pulse is frequency chirped.

II. Interferometric autocorrelation

Unlike intensity autocorrelations which average out phase information, interferometric autocorrelations produce unique patterns depending on the degree of chirp in the laser pulse. The normalized second order interferometric autocorrelation function \( A_2(\tau) \) is given by the expression [8]:

\[
A_2(\tau) = \frac{\int_{-\infty}^{\infty} \left| \left\{ E(t) + E(t-\tau) \right\} \right|^2 \, dt}{2 \int_{-\infty}^{\infty} |E(t)|^4 \, dt}
\]

where \( E(t) = E_0(t) \exp\{ i(\omega t + \phi(t)) \} \), \( \phi \) is the phase, and \( \tau \) is the delay time. At zero delay, constructive interference between each pulse yields an amplitude of 16 for \( A_2(\tau) \). When the delay between the two pulses is increased by \( \lambda/2 \), destructive interference occurs and the resulting amplitude is nearly zero. For much longer delay times, the envelopes of
constructive and destructive interference join and have a background value of 2. The characteristic peak to background ratios for second and third order interferometric and intensity autocorrelations is given in Table 4.1.

<table>
<thead>
<tr>
<th></th>
<th>2nd order</th>
<th>3rd order</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intensity</td>
<td>3:1</td>
<td>10:1</td>
</tr>
<tr>
<td>Interferometric</td>
<td>8:1</td>
<td>32:1</td>
</tr>
</tbody>
</table>

The shape of the interferometric autocorrelation function is dependent on the type of frequency chirp in the laser pulse. For a linearly chirped Gaussian pulse, the electric field can be written as:

\[ E(t) = \exp \left( -\frac{t^2}{2T^2} \right) \exp \left( i \left( \omega_0 t + \frac{a\tau^2}{T^2} \right) \right) \]

where \( T \) is the pulse width and \( a \) is the chirp parameter. The second order interferometric autocorrelation function \( A_2(\tau) \) of a linearly chirped Gaussian pulse is given by:

\[ A_2(\tau) = 1 + 2\exp \left( -\frac{\tau^2}{2T^2} \right) + \exp \left( -\frac{\tau^2}{2T^2} - \frac{2a\tau^2}{T^2} \right) \cos(2\omega_0 \tau) \]

\[ + 4\exp \left( -\frac{3\tau^2}{8T^2} - \frac{a^2\tau^2}{2T^2} \right) \cos \left( \frac{a\tau^2}{2T^2} \right) \cos(\omega_0 \tau) \]

The dependence of the shape of the autocorrelation function \( A_2(\tau) \) on the degree of chirp is shown in Figure 4.1. As the degree of chirp increases, the depth of modulation decreases and the time interval over which interference occurs decreases. When the degree of chirp is large, the beginning and end of the pulse are no longer coherent and, thus, interference effects are lost. In the case of a strongly chirped pulse, the interferometric autocorrelation
Figure 4.1: Dependence of the second order interferometric autocorrelation function on the degree of chirp of a linearly chirped Gaussian pulse with electric field \( E = \exp\{ -0.5(t/T)^2 \} \exp\{ i(\omega t + a(t/T)^2) \} \). For a T=500 fs pulse width and \( \omega=7.5e15 \) s\(^{-1}\), the chirp parameter for each individual plot is a) \( a=0 \), b) \( a=1 \), c) \( a=2 \), and d) \( a=5 \).
function becomes similar to the normal intensity autocorrelation function. The normal second order intensity autocorrelation function \( a_2(\tau) \) is given by the time average of the interferometric autocorrelation function:

\[
a_2(\tau) = \langle A_2(\tau) \rangle = 1 + 2\exp\left( -\frac{\tau^2}{2T^2} \right)
\]

since \( \langle \cos(2\omega \tau) \rangle = \langle \cos(\omega \tau) \rangle = 0 \).

The same analysis can also be carried out for third order autocorrelation functions. Given the normalized third order interferometric autocorrelation function:

\[
A_3(\tau) = \frac{\int_{-\infty}^{\infty} \left| \langle E(\tau) + E(t-\tau) \rangle \rangle^3 \right|^2 dt}{2 \int_{-\infty}^{\infty} |E(t)|^6 dt}
\]

and a linearly chirped chirped Gaussian pulse, the autocorrelation function \( A_3(\tau) \) is given by:

\[
A_3(\tau) = 1 + 9\exp\left( -\frac{2\tau^2}{3T^2} \right) + 9\exp\left( -\frac{3\tau^2}{4T^2} - \frac{a_2^2 \tau^2}{3T^2} \right) \cos(\omega \tau)
\]

\[
+ 6\exp\left( -\frac{5\tau^2}{12T^2} - \frac{a_2^2 \tau^2}{3T^2} \right) \cos\left( \frac{2a_2^2 \tau^2}{3T^2} \right) \cos(\omega \tau)
\]

\[
+ 6\exp\left( -\frac{2\tau^2}{3T^2} - \frac{4a_2^2 \tau^2}{3T^2} \right) \cos\left( \frac{2a_2^2 \tau^2}{3T^2} \right) \cos(2\omega \tau)
\]

\[
+ \exp\left( -\frac{3\tau^2}{4T^2} - \frac{3a_2^2 \tau^2}{T^2} \right) \cos(3\omega \tau)
\]

Figure 4.2 shows how the degree of chirp affects \( A_3(\tau) \). As for the second order case, the interferometric autocorrelation function becomes similar to the intensity autocorrelation
Figure 4.2: Dependence of the third order interferometric autocorrelation function on the degree of chirp of a linearly chirped Gaussian pulse with electric field \( E = \exp\{ -0.5(\frac{t}{T})^2 \} \exp\{ i(\omega t + a(\frac{t}{T})^2) \} \). For a T=500 fs pulse width and \( \omega=7.5e15s^{-1} \), the chirp parameter for each individual plot is a) a=0, b) a=1, c) a=10, and d) a=100.
function when the degree of chirp is strong. The third order intensity autocorrelation function $a_3(\tau)$ is given by:

$$a_3(\tau) = \langle A_3(\tau) \rangle = 1 + 9\exp \left( -\frac{2\tau^2}{3T^2} \right)$$

Figure 4.1 and 4.2 also illustrate the peak to background ratios for each order autocorrelation function. By using the spectrum, intensity autocorrelation, and interferometric autocorrelation, the amplitude and phase of the laser pulse can be determined.

III. Single shot phase sensitive autocorrelation

Szabo et al have developed a single shot phase sensitive autocorrelator (PSA) [4] that is similar to the multiple shot interferometric technique previously discussed. In contrast to the interferometric autocorrelation technique, the PSA allows the chirp of a single laser pulse to be determined and does not require sub-wavelength accuracy for controlling the length of the delay line. The intensity autocorrelation technique presented in Chapter 2 and the PSA are both based on single shot autocorrelation using tilted pulse front geometry. Tilted pulse front autocorrelation can be created by combining two non-collinear beams at an angle (Figure 3.4) or by combining two collinear beams which have their pulse fronts tilted by a dispersive element. When the tilted pulse fronts overlap in a nonlinear medium, the autocorrelation trace is created along the bisector of the pulse fronts [4].

Figure 4.3 shows the experimental arrangement for the PSA for ultrafast visible laser pulses. The device consists of a modified Michelson interferometer containing prisms in each arm of the interferometer to tilt the pulse front of each copy of the laser pulse. The pulse front delay is created due to the difference between the phase velocity and group velocity in the dispersive prism [11]. The angle $\gamma$ of the pulse front delay is given by
Figure 4.3. Experimental setup for a single shot phase sensitive autocorrelator for ultrafast visible laser pulses. A prism in each arm of the interferometer causes the pulse front of the original pulse to be tilted by an angle $\gamma$. When the two pulse fronts overlap spatially and temporally, second harmonic light is generated by the nonlinear crystal. The mirrors in each arm of the interferometer are slightly misaligned to create a vertical fringe pattern in the output beams.
\[ \tan \gamma = \lambda \frac{d\varepsilon}{d\lambda} \] where \( d\varepsilon/d\lambda \) is the angular dispersion of the prism and \( \lambda \) is the wavelength of the laser. Expressing the angular dispersion in terms of the geometry of the prism, the expression for the pulse front tilt angle \( \gamma \) can be written as:

\[
\tan \gamma = \lambda \frac{dn}{d\lambda} \frac{nsin\alpha}{\sqrt{n^2 - (\sin \varepsilon)^2}} \sqrt{1 - \left( \cos \alpha \sin \varepsilon - \sin \alpha \sqrt{n^2 - (\sin \varepsilon)^2} \right)^2}
\]

where \( n \) is the index of refraction of the prism, \( \alpha \) the apex angle of the prism, \( dn/d\lambda \) the dispersion of the prism material, and \( \varepsilon \) the angle of incidence on the prism. The two beams exiting the interferometer are slightly misaligned in order to produce a vertical interference fringe pattern in the output beams. The two beams are focused by a cylindrical lens on a second harmonic generating nonlinear crystal. The image of the second harmonic light in the crystal is then detected by a spatial detector. The spatial distribution of the second harmonic light produces a second order phase sensitive autocorrelation function. The spatial axis of the autocorrelation trace is converted to a time axis by introducing a known delay \( \Delta T \) in one arm of the interferometer and observing the shift \( \Delta X \) in the peak of the autocorrelation trace. The width \( S \) of the spatial distribution of the second harmonic light is related to the temporal width \( A \) of the autocorrelation function by \( A = S \Delta T / \Delta X \). With this type of experimental setup, the overlap of the two pulses emerging from the interferometer is defined by the tilt angle \( \gamma \) induced by the prisms and the pulse width. Figure 4.4 shows how the length of the overlap region is influenced by these parameters.

According to reference [4], the time integrated intensity of the second order phase sensitive autocorrelation trace is given by:

\[ I_{SH} = \int_{-\infty}^{\infty} \left( E_1(t) + E_2(t-x) \right)^4 dt \]

where \( E_1(t) \) and \( E_2(t-x) \) are the electric fields of the pulses emerging from each arm of the autocorrelator. Assuming a linearly chirped Gaussian pulse, the expressions for the electric fields are:
\[ z = \frac{c\Delta t}{2n} \]
\[ y = \frac{z}{\cos \gamma} \]
\[ x = \frac{y}{\tan \gamma} = \frac{c\Delta t}{2nsin\gamma} \]

Figure 4.4 The overlap region of two pulses with tilted pulse fronts is a function of the tilt angle \( \gamma \), the pulse width \( \Delta t \), and the index of refraction \( n \). The length \( d \) of the overlap region is given by \( d = 2x = c\Delta t/nsin\gamma \).
\[ E_1(x,t) = A(x) \exp \left\{ -2 \ln 2 \left[ \frac{t - 2xt\gamma/c}{\tau} \right]^2 \right\} \exp \left\{ i \left( \omega t + at\left[ t + 2xt\gamma/c \right] - k_1 x \right) \right\} + c.c. \]

\[ E_2(x,t) = A(x) \exp \left\{ -2 \ln 2 \left[ \frac{t - 2xt\gamma/c}{\tau} \right]^2 \right\} \exp \left\{ i \left( \omega t - at\left[ t - 2xt\gamma/c \right] - k_2 x \right) \right\} + c.c. \]

where \( A \) is the electric field amplitude, \( \gamma \) is the pulse tilt angle after one transit through a prism, \( \tau \) is the pulse duration, \( c \) is the speed of light, \( k_{1x} \) and \( k_{2x} \) are the \( x \) components of the wave vectors, \( a \) is the chirp parameter, and c.c. denotes the complex conjugate part.

By fitting a calculated autocorrelation trace to an experimentally observed trace, the chirp parameter \( a \) can be determined. The degree of chirp effects the shape of the autocorrelation trace in the same way that the degree of chirp influences the shape of the interferometric autocorrelation trace.

In order to use the PSA for ultraviolet laser pulses, two components of the experimental arrangement must be modified. First, the apex angle and material of the prisms must be chosen to obtain the proper angle \( \gamma \) for the pulse front delay. Because the dispersion of most materials in the ultraviolet is much larger than in the visible, a prism used for visible lasers will create too large of a tilt angle \( \gamma \) for ultraviolet lasers. Table 4.2 shows the first and second order dispersion of fused silica and calcium fluoride at four ultraviolet excimer laser wavelengths. Secondly, the nonlinear crystal must be replaced by a nonlinear medium that generates second or third order autocorrelation traces for ultraviolet lasers.

For single shot phase sensitive autocorrelation of the subpicosecond KrF excimer laser, fluorescence of the self trapped exciton in calcium fluoride is used as the nonlinear medium. Using this fluorescence for single shot autocorrelation of a subpicosecond KrF laser pulse, Hata et al [14] have demonstrated that the exciton fluorescence depends quadratically on the incident KrF laser intensity. The wavelength of the peak fluorescence intensity is centered at 300 nm. For the length \( d \) of the overlap region (see Figure 4.4) in
### Dispersion data for fused silica

<table>
<thead>
<tr>
<th>wavelength [(\mu m)]</th>
<th>index of refraction</th>
<th>(-\frac{dn}{d\lambda}) [(\mu m^{-1})]</th>
<th>(\frac{d^2n}{d\lambda^2}) [(\mu m^{-2})]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.351</td>
<td>1.4767</td>
<td>0.1662</td>
<td>1.5545</td>
</tr>
<tr>
<td>0.308</td>
<td>1.4856</td>
<td>0.2576</td>
<td>2.8682</td>
</tr>
<tr>
<td>0.248</td>
<td>1.5086</td>
<td>0.5608</td>
<td>8.5578</td>
</tr>
<tr>
<td>0.193</td>
<td>1.5608</td>
<td>1.5906</td>
<td>38.4825</td>
</tr>
</tbody>
</table>

### Dispersion data for calcium fluoride

<table>
<thead>
<tr>
<th>wavelength [(\mu m)]</th>
<th>index of refraction</th>
<th>(-\frac{dn}{d\lambda}) [(\mu m^{-1})]</th>
<th>(\frac{d^2n}{d\lambda^2}) [(\mu m^{-2})]</th>
</tr>
</thead>
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<tr>
<td>0.351</td>
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<td>1.0596</td>
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<tr>
<td>0.308</td>
<td>1.4526</td>
<td>0.1764</td>
<td>1.9167</td>
</tr>
<tr>
<td>0.248</td>
<td>1.4680</td>
<td>0.3735</td>
<td>5.4208</td>
</tr>
<tr>
<td>0.193</td>
<td>1.5015</td>
<td>0.9799</td>
<td>21.1694</td>
</tr>
</tbody>
</table>

**Table 4.2** Dispersion data for fused silica and calcium fluoride are presented in the table above. The first and second order dispersion are calculated from the index of refraction polynomials given in [12] and [13].
calcium fluoride to be \( \sim 1 \) cm, the pulse front tilt angle \( \gamma \) for a 500 fs pulse should be 0.55° after two passes though a prism.

IV. Experimental setup and preliminary results

The experimental setup of the phase sensitive autocorrelator for the subpicosecond KrF excimer laser is shown in Figure 4.5. Two copies of the incident laser pulse are produced by a 50% beamsplitter. Fused silica wedged windows with wedge angles of 2° are used as prisms to tilt the pulse front. A wedge angle of 2° produces a pulse front tilt angle of 0.55° after two passes through a prism. The two beams emerging from the interferometer are focused by a cylindrical lens (\( f=10 \) cm) to the center of a calcium fluoride window. The exciton fluorescence from the calcium fluoride window is imaged onto a Cohu CCD camera by a series of spherical lenses. The CCD camera is located above the plane of the autocorrelator because the autocorrelation trace is obtained by viewing the exciton fluorescence pattern perpendicular to the propagation direction of the output beams from the interferometer and perpendicular to the plane of the autocorrelator. The fluorescence image detected by the CCD camera is shown in Figure 4.6. From the profile of this image along the axis of greatest intensity, the single shot phase sensitive autocorrelation trace is obtained. As discussed in Chapter 3, the time scale of the autocorrelation is obtained by noting the number of pixels the maximum of the fluorescence pattern moves for a given delay introduced by a translation stage. For the present detection system, the calibration factor is approximately 11 fs/pixel. The translational stage used in the delay line has a resolution of 2 \( \mu \)m.

Preferably, a camera lens should be used to image the fluorescence onto the CCD camera. However, the material used for most camera lenses does not transmit ultraviolet light near 300 nm. Because single element calcium fluoride lenses are used to image the
Figure 4.5. Schematic diagram for single shot phase sensitive autocorrelation of the subpicosecond KrF excimer laser pulse. Fused silica wedged windows with a wedge angle of 2° are used to tilt the pulse front of the input pulse. When the two tilted pulse fronts overlap spatially and temporally at the focus of the cylindrical lens, exciton fluorescence in the calcium fluoride window yields the autocorrelation trace. The fluorescence is detected by a CCD camera located perpendicular to the plane of the paper.
Figure 4.6. False color image of the calcium fluoride exciton fluorescence detected by the CCD camera. The image is distorted (the outer edges are not in focus) because of spherical aberration in the lenses used to image the fluorescence on the CCD camera. The autocorrelation trace in Figure 4.7 is obtained by making a horizontal profile along the axis of greatest intensity.
fluorescence onto the CCD camera, the camera detects an image that is distorted by spherical aberration. Figure 4.7 shows the autocorrelation trace obtained from the raw image in Figure 4.6. If the distorted image can be corrected by assuming a Gaussian shaped distortion, the autocorrelation trace becomes as shown in Figure 4.8.

In principle, the XeF gas cell that is used for the autocorrelation method presented in Chapter 2 can also be used as the nonlinear medium for the single shot phase sensitive autocorrelator. The XeF gas cell was initially used for the experiment; however, the XeF fluorescence intensity was too weak to be detected by the present detection system. In order to use the XeF cell for the nonlinear medium, the appropriate apex angle for the prisms would be ~ 3°. In fused silica, this yields a pulse tilt angle of 0.83° after the beam passes twice through a prism, and the length d of the region of overlap is 1 cm.
Figure 4.7. Autocorrelation trace obtained from the image in Figure 4.6.
Figure 4.8. Phase sensitive autocorrelation trace obtained by correcting the curve in Figure 4.7. A Gaussian shaped curve was fitted to the curved background in Figure 4.7 so that the horizontal background level could be extracted.
References


Conclusion

The goal of this work was to develop diagnostic procedures for characterizing a subpicosecond KrF excimer laser system. To this end, a one meter spectrometer, a single shot intensity autocorrelator and a single shot phase sensitive autocorrelator were constructed. Spectral analysis with the one meter spectrometer was shown to be a simple and sensitive method for aiding in the daily alignment of the laser system. From experimental observations of the UV laser spectrum, a model was devised to explain the shape of the UV laser spectrum. Near the center of the KrF gain bandwidth, the spectral shape of the laser is shown to be influenced by self phase modulation. From the self phase modulation model, it has been shown that the UV laser pulse has an asymmetric pulse shape and a linear component of phase shift of $\sim 5\pi$ during the 500 fs pulse width. The behavior of the laser spectrum as it is tuned near the long wavelength side of the gain is influenced by resonance with the $v=1$ and $v=2$ vibrational levels of the KrF upper state.

The pulse width of the UV laser was measured by a single shot intensity autocorrelation technique based on the spatial distribution of XeF fluorescence in a pressure cell. A pulse width of 477 fs was measured with this third order correlation technique. In order to determine the frequency chirp of the laser pulse, a single shot phase sensitive autocorrelator based on exciton fluorescence in calcium fluoride was designed. The chirp of the laser pulse can be obtained by fitting calculated autocorrelation traces to the observed autocorrelation traces.

Two important tasks remain in order to complete the work presented in this thesis. First, the detection of the calcium fluoride exciton fluorescence must be improved to obtain phase sensitive autocorrelation traces that are free of image distortion. Secondly, a computer program must be written to calculate the chirp of the laser pulse from the experimentally observed phase sensitive autocorrelation traces. Once these tasks are
completed, the degree of frequency chirp determined with the phase sensitive autocorrelator will be correlated with the shape of the UV laser spectrum and compared with the chirp parameters calculated with the self phase modulation model.

From a practical point of view, this thesis is also meant to serve as a comprehensive training manual for teaching new graduate students how to operate and maintain the laser system.
Appendix A

Ultrashort Pulse KrF Excimer Laser System Manual
Ultrashort Pulse KrF Excimer Laser System Manual

by

Shoichi Kubodera, Stephen Paul Le Blanc
and Roland Sauerbrey

Department of Electrical and Computer Engineering
Rice University
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   A.6 1-m spectrometer for the analysis of laser spectra
   A.7 Polarization of the laser beam
I. Introduction

This manual describes the main features of the operation of an ultrashort pulse excimer laser system shown in Figure A1.

The experiences of the authors with the ultrashort pulse excimer laser system are described. It is not a substitute for a general excimer laser manual which is available from the excimer laser manufacture (Lambda Physik), but rather focuses on the specific features of the dye laser system as well as the interplay of the dye and excimer lasers. The manual in its present form represents our knowledge after about one year of operation. We intend to improve the manual periodically and incorporate new experiences. For this purpose, we hope to obtain input from the manufactures of this system as well as from other users.
Figure A1. Physical diagram of the laser system
Figure A1. (continued) Physical diagram of the laser system
II. Brief Description of the System

This ultrashort pulse excimer laser system is shown in Figure A1. It consists of a double chamber excimer laser (EMG-150, Lambda Physik) and an ultrashort pulse dye laser system (Laser Laboratorium Göttingen). The XeCl laser beam from one chamber of the excimer laser is used to pump a sequence of dye lasers. This sequence of dye lasers produces a subpicosecond green (496 nm) laser pulse that is frequency doubled by means of a nonlinear crystal (BBO). The doubled frequency beam is amplified twice in the KrF laser medium.

The quenched cavity dye laser (QCDL, PTP dye in cyclohexane) is pumped by a 15 ns XeCl laser pulse. The off-axis high-Q resonator of the QCDL efficiently quenches the on-axis oscillation in a low-Q cavity and the on-axis output of the QCDL has a pulse width of ~ 200 ps at 340 nm. The output of the QCDL longitudinally pumps the short cavity dye laser (SCDL) which is a 0.5 mm long cavity filled with B-PBD dye in methanol. This SCDL provides rapid feedback for laser oscillation and produces a ~ 15 ps pulse at 365 nm. After amplification of this 365 nm beam (B-PBD in ethanol) the beam passes through the gated saturable absorber (GSA) which tailors the tail of the pulse to produce a ~ 9 ps pulse. The amplified GSA output pumps the distributed feedback dye laser (DFDL, C307 dye in DMF) which is heart of this system. Distributed feedback operation is accomplished by using a transmission grating and a microscope objective to produce a periodic interference pattern on the DFDL dye cell. The gain modulation in the cell leads to distributed feedback operation and produces a subpicosecond pulse. The DFDL output provides a pulse with a pulse width of ~ 0.5 ps tuned at 497 nm. This green beam is amplified twice in a conventional side-pumped amplifier (C102 dye in ethanol) and in a Bethune cell (C307 in ethanol) to increase the energy to ~ 20 μJ. A nonlinear crystal (BBO)
produces UV radiation at 248 nm. This subpicosecond UV laser beam with an output energy of ~ 2 μJ is introduced to the KrF excimer laser amplifier. A vacuum spatial filter is located between the first and second pass through the KrF amplifier to minimize the ASE contribution to the final output. The KrF laser amplified beam after the spatial filter is expanded to fill the KrF gain medium and amplified in the saturated regime. The final output of the KrF laser provides an energy of ~ 15 mJ in ~ 0.5 ps at 248 nm.
Ultrashort Pulse Excimer Laser System Manual

A diagram of the femtosecond laser system can be found in Figure A1. The alignment instructions that follow refer to optical elements that are labeled and defined in the diagram.

III. Comprehensive Alignment Procedure

The comprehensive alignment procedure describes the alignment of the laser system from the point of installation. The steps outlined below should be followed only when some portion of the laser system has been severely misaligned. The more frequent type of maintenance procedures and daily alignment routines follow this section.

The following preparations/modifications of the original laser system have been made:

a) Gas handling system of EMG-150 excimer laser: To avoid mixing different halogen gases (F₂ and HCl), a separate gas handling system has been installed inside the excimer laser body. The parts required for this modification are two gas pressure gauges, three solenoid valves, switches, a vacuum valve and some metric tubing.

b) Interlock system: The excimer laser power supply is connected with an interlock which is activated by the dye circulators' power supply. See EMG-150 manual for detail of the excimer interlock system.

c) Pulse counter for the excimer laser: A pulse counter with an eight digit display counts each pulse.
d) A separate 5 volt power supply for the motor that rotates the OG 530 saturable absorber has been installed.

When the laser system arrives:

- Release transportation locks
- Inspect all optical elements used in the system
- Check the BBO crystal orientation
  
  - The BBO crystal is properly oriented if the height of the crystal is slightly larger than the width. See Figure A2.

![Figure A2: Orientation of the BBO crystal](image)

1. Dye circulators and motor for saturable absorber rotation
   
   - Power (3 - 5 V\textsubscript{DC}) should be switched on before excimer is turned on.
   - If the dye circulators have not been turned on, then the interlock switch will prohibit the high voltage of the excimer laser from being activated.

2. Inspect the condition of the dye solutions.
   
   - Dye reservoirs should be filled to the appropriate level. If the reservoir level is low, then add solvent until the appropriate level is reached. (Cyclohexane tends to evaporate faster than the other solvents.)
   - Replace dye solutions according to the schedule in Appendix A.3.
- Record the number on the pulse counter whenever a dye solution or dye filter is replaced.

3. Check the XeCl resonator alignment

- Use the He-Ne beam and circular apertures to define the XeCl resonator axis.
- Use back reflections of the He-Ne beam incident on the oscillator mirrors to judge whether the oscillator mirrors are perpendicular to the resonator axis.
- Never move the He-Ne laser tube and the mirror MO.

![Figure A3: Alignment of the XeCl laser tube with the He-Ne laser](image)

4. Check the XeCl laser energy

- Attenuate the XeCl laser energy to ~ 60 mJ by introducing thin fused silica plate(s) in the path of the XeCl pump beam between the mirrors M1 and M2. (This will cause a slight shift of the XeCl pump beam with respect to all beam splitters.)
5. Alignment of the XeCl pump beam through dye laser beam splitters
   - Center the XeCl pump beam on the mirror M2 by moving the mirror M1.
   - Examine the far-field pattern of the pump beam at the Bethune cell (H). (It should be symmetrical. If not, move the mirror M2.)
   - The pump beam should be centered on cylindrical lenses (CL7) in front of Bethune cell.
   - After this alignment, check the horizontal position of the pump beam on each dye cell. If the positions are away, then adjust all beam splitters (BS1-3) and the mirror M8.

6. Observation of the beam pattern from the QCDL (A)
   - If the spot is not centered on the pinhole P1, then rotate the dye cell (A) for horizontal adjustment and/or move the mirror M4 for vertical adjustment.

7. Quenched operation of the QCDL (1)
   Note: When blocking the quenching mirror M3, always introduce a beam block before the SCDL to avoid too high of an intensity on the SCDL cell.
   - Align the quenching mirror M3 parallel with the resonator formed by the dye cell (A) to get one bright spot (in line resonator). See Figure A4.
   - Then as viewed from the top, turn the mirror M3 counter clockwise until the characteristic beam pattern emerges.
   - Check that the quenched spot is centered on the pinhole P1. If not, then go to Step 6.
8. Quenched operation of the QCDL (2)

- By turning the quenching mirror M3, minimize the brightness of the dye laser beam before the first BPBD amplifier (C) to get optimal quenched operation.

9. Optimization of the GSA output

- The beam pattern after the GSA consists of a very sharp spot and a diffused part.

Figure A5: The beam pattern of the GSA output

- Optimize the brightness of the sharp transmitted spot by turning the mirror M5.
10. Alignment of the beam through the pinhole P4

i) Horizontal alignment

- Adjust the second BPBD amplifier output for maximum brightness by moving up or down the cylindrical lens (CL3) in front of the second BPBD amplifier (E).
- If the beam is not in the middle of the pinhole P4, then obtain a new vertical position of the cylindrical lens (CL2) in front of the first BPBD amplifier (C). Move the cylindrical lens CL2 down in order to move the beam to the right, or move the cylindrical lens CL2 up to move the beam to the left.
- Make sure that the cylindrical lenses are parallel to the dye cells.
- Go to step 9 until the beam is horizontally aligned.

Note: For very precise alignment of the beam in the horizontal direction, adjust the beam splitter (BS2) for the second amplifier (E) instead of moving the cylindrical lenses because it is difficult to adjust the cylindrical lenses very precisely for fine alignment. Always check the second BPBD amplifier first. (You may not have to follow the procedure!)

ii) Vertical misalignment

- Adjust the second BPBD amplifier cell (E) to give brightest output.
- If the beam is not in the middle of the pinhole P4 then move the first BPBD cell (C) to a new position. Move the dye cell (C) backward to move the beam down, or move the dye cell (C) forward to move the beam up.
- Go to step 9 until the beam is vertically aligned.
- If you get a nearly optimum beam pattern after the second BPBD amplifier (E), then you can adjust the second dye amplifier cell (E) to obtain the optimum beam pattern on the T-shape beam stop in the microscope objective. See Figure A6.
(Make sure that you still have the optimum beam pattern after the second amplifier after this adjustment.)

![Diagram of 1st and 0th order diffraction]

**Figure A6:** Optimum diffracted beam pattern on the T-shape beam stop

11. Alignment of the DFDL (If the DFDL is lasing, congratulations, skip to step 12.)

- Objective translation with coarse and fine micrometers.
- The microscope objective in front of the DFDL cell has a limited range of movement. The tip of the microscope objective should not be moved too far away from the DFDL cell or the immersion oil spot will separate.
- If the objective is moved too close to the DFDL cell, then the objective will be exerting too much pressure on the dye cell.
This gap will start to shrink when the objective is too close to the DFDL cell.

![Diagram of microscope objective and DFDL cell](image)

**Figure A7:** The microscope objective and the DFDL cell

- Move the objective translational stage (CCW rotation of the micrometer) towards the DFDL cell until the gap on the microscope objective first starts to shrink.
- Then move the objective translational stage (CW rotation of the micrometer) away from the cell until the DFDL operates at the maximum brightness.

*Note: DFDL intensity varies with translation of the objective. See Figure A8.*

![Graph of intensity vs. translation](image)

**Figure A8:** Intensity variation by objective translation

- The fine adjustment screw for the objective translation can be adjusted later to give the optimal output of the DFDL.

12. Pumping level of the DFDL
   - The DFDL should be pumped at twice threshold to get stable single-pulse
operation.
- Place a $T = 0.5$ ND filter in front of the T-shape beam stop in the DFDL pump beam. Always tilt the ND filter to avoid unnecessary feedback in the system.
- Increase/decrease the pumping energy of the DFDL with the beam block BB1 until the threshold is just reached for the DFDL operation.
- Remove the ND filter.

13. Centering of the DFDL output beam on the pinhole P5
   - Check whether the DFDL output beam is centered on the pinhole P5. If not, adjust the micro-translational stage of the pinhole P5 both vertically and horizontally.

14. Centering of the DFDL output beam on the pinhole P6
   - Check whether the beam is centered on the pinhole P6.
   - For vertical movement: move the mirror M7 and the beam splitter BS3 for the C102 amplifier (G).
   - For horizontal movement: move the mirror M7 and translate the C102 dye cell (G) back and forth.

15. Check the rotation of the saturable absorber
   - Sometimes the glass filter saturable absorber stops rotating. Check the rotation occasionally to avoid damaging the filter.

16. Alignment of the Bethune cell'
   - Block the XeCl pump beam at M8 and determine if the dye laser beam is coaxial with the capillary tube. If not, adjust the x-y translational stage of the Bethune
cell.
- When the cell is pumped, a homogeneous beam pattern from the Bethune cell should be observed. If not, check the vertical alignment of the cylindrical lenses (CL7) in front of the Bethune cell. Make sure that the cylindrical lenses (CL7) are parallel to the Bethune cell after this adjustment.

17. Alignment of the beam through the BBO crystal
   - A uniform diffraction pattern of the green dye laser beam should be observed in front of the BBO cell. If not, then go to the step 16.
   - If the green beam is not hitting the crystal, move the mirror M10.

18. BBO crystal phase matching
   - Adjust the angle of the BBO crystal for efficient second harmonic generation.
   - When optimizing the BBO angle, the best place to observe the second harmonic beam is just before the mirror M13.
   - The horizontal adjustment is more sensitive for second harmonic generation than the vertical adjustment.
   - The path of the second harmonic beam depends on the way the BBO crystal is situated in the cell. The fundamental and second harmonic beams may appear coincident when the two beams are viewed close to the BBO crystal.

19. Take a rest.

20. Alignment of the 248 nm beam (1)
   - The second harmonic beam (248 nm) should hit the center of the half-cut mirror M13. If not, move the mirrors M11 and M12.
21. Alignment of the 248 nm beam (2)

- The first amplified beam exiting from the KrF discharge tube should hit the middle of the mirror M14.

- Ideally, the seed beam is just clipped at the top by the top edge of the KrF tube aperture and just clipped at the bottom by the mirror M17. See Figure A9.

Figure A9: The first amplified KrF beam pattern in the ASE background

- It may be better to use a diluted KrF gas mixture (less F₂) to discriminate this beam pattern from the ASE background.

- The excimer laser may need to be rotated with respect to the He-Ne beam axis to optimize the first amplified beam path.

- If the amplified beam is not observed, check the seed beam alignment again. If the seed beam is still not detected, then move the transmission grating to tune the second harmonic wavelength in the KrF gain region.

- If the amplified beam is not observed yet after tuning, then check the timing of the firing between the XeCl and KrF laser tubes. The appropriate time difference is 30 -40 ns.

Note: When aligning the seed beam, it may be helpful to stop the firing of the KrF discharge tube. The EMG-150 we have has a switch which allows the XeCl tube to discharge without firing the KrF tube.
22. Alignment of the beam through the vacuum spatial filter (I)
   - Obtain a homogeneous beam profile after the beam telescope (L9 and L8) by
     adjusting the x-y-z stage of the pinhole cell and by turning the mirror M10 for
     fine adjustment.
   - The pressure in the vacuum spatial filter must be checked and evacuated down to
     \(10^{-3}\) Torr once a week. The spattered pinhole material (fused silica) may increase
     the pressure inside the spatial filter.
   - During operation, red fluorescence from the bottom window of this cell will be
     observed.

--------------------Spectrometer is required for the following procedures--------------------

23. Spectrographic analysis for fine optimization
   i) DFDL pump level
      - Operate at the highest possible pump level while still remaining in the single pulse
        regime. Ripples or small modulations appear in the KrF spectrum when this
        occurs.
      - Alternatively, check the spectral shape of the green beam. This should have a
        smooth, one-peak shape.
   ii) Translation of the microscope objective and the transmission grating
      - When the transmission grating and the objective translational stage are translated
        back and forth simultaneously, so that the peak of the spectrum does not shift, an
        optimum can be found for broadest spectrum and maximum amplitude. This
means the pump region is moved back and forth in the DFDL cell (pump depth \( \sim 5 - 10 \, \mu m \)).

- When the transmission grating is only moved back and forth, the spectral shape should change smoothly. If not, check the objective translational stage to optimize the DFDL output.

![Optimum Intensity vs Wavelength](image)

Figure A10: Change of the KrF spectral shape by translating the transmission grating

iii) Adjustment of the saturation level of the Bethune cell amplifier

- In order to obtain a smooth, double peaked spectral shape for the KrF beam, adjust the saturation level of the Bethune cell with the beam block BB2. This is important for obtaining a linearly chirped pulse.

24. Optimization of the XeCl and KrF gas mixtures

- In order to adjust the timing between the oscillator (XeCl) and the amplifier (KrF), observe the temporal shape of the ASE from the KrF discharge tube. The ASE will be depleted by the saturated second amplification. The timing of the second amplified beam should be just after the ASE peak. (The timing of the first amplified beam will be just before the peak of the ASE under this condition.) See
Figure A11. (Appendix A.5 provides more detail.)

Depleted ASE due to the second amplification

Figure A11: Typical temporal profile of the ASE background with femtosecond laser injection

- If possible use less F₂ concentration for the KrF gas mixture to minimize the ASE contribution in the laser beam.
IV. Periodic Maintenance Check List

1. Quenching of QCDL (PTP)
   - Adjust the mirror M3 to minimize the beam spot brightness at M5 for the most quenched operation. (See step 7 in the comprehensive alignment procedure.)

2. Optimization of GSA output
   - Adjust the mirror M5 until the characteristic sharp beam spot is observed before the dye cell (E). (See step 9 in the comprehensive alignment procedure.)

3. Translation of DFDL microscope objective
   - See step 11 in the comprehensive alignment procedure.

4. DFDL output centering
   - Check if the DFDL beam is coming through the pinhole P5. If not, adjust the pinhole translational stage (x-y) so that the green beam passes through.

5. Beam centering before saturable absorber (color glass plate)
   - Check if the amplified DFDL beam is coming through the pinhole P6. If not, alternately adjust the mirror M5 and the dye cell (C) until the beam is through the pinhole P6.

6. Positioning of cylindrical lens assembly (CL7) in front of Bethune cell
   - Move the lens assembly vertically to get a uniform beam pattern on the Bethune cell.
7. BBO phase matching
   - Adjust the horizontal and vertical angles to get optimum second harmonic output.
     The horizontal angle adjustment is more sensitive.

8. Evacuation of vacuum spatial filter
   - Evacuate the vacuum spatial filter (I) down to $10^{-3}$ Torr once a week.

V. Everyday Check List

1. Check the dye conditions
   - All dye reservoirs should be filled to the appropriate level. If some solvent has evaporated from a reservoir, fill the reservoir with the appropriate solvent.
   - Dye circulation pumps should be switched on before the excimer is turned on.

2. Check the XeCl laser energy
   - Set the XeCl laser energy ~ 60 mJ using the appropriate number of fused silica plate(s) to attenuate the XeCl output beam.
   - Check the energy value during the operation.

3. Check the DFDL pump energy
   - Set the pump level of the DFDL to two times above threshold using a $T = 0.5$ ND filter.
   - Recheck this pump level after the laser system warms up. A change in temperature will cause a shift in the laser wavelength and the beam directionality.
4. Align the Bethune cell

- Adjust the x-y translational stage of the Bethune cell until a uniform green dye laser beam pattern is observed. (Block the XeCl pump beam of the Bethune cell when performing this.)

5. Align the UV beam through the vacuum spatial filter

- Check the uniformity of the expanded KrF laser beam after the vacuum spatial filter.
- Adjust the x-y-z translational stage of the pinhole and alternately adjust the mirrors M9 and M10 for more precise alignment of the KrF seed beam through the pinhole.

Spectrometer is required for the following procedure.

6. Tune the DFDL

- Translate the transmission grating before the DFDL cell to tune the green wavelength appropriately. (Note: This is a very sensitive adjustment, requiring the use of the single-shot 1m-spectrometer.)
- Adjust the BBO cell to get optimum phase matching after this translation.

7. Adjust the saturation level of the C307 dye laser

- Change the pump level of the Bethune cell or insert a ND filter before the 2nd amplification of the KrF laser beam. The KrF output spectrum should have a smooth double peaked shape.
  - A low saturation level will cause a single peak in the KrF spectrum.
Appendix A.1: Beam patterns at various positions in the system

(1) at P1

with M3

~ 0.5 mm

without M3

(2) at M5 (after P2)

~ 1 mm

(3) at 1st amp BPBD

1 ~ 2 mm

(4) at GSA

~ 0.3 mm

(5) at 2nd amp BPBD
diffused

very sharp

(6) at CL4

~ φ 2 mm

(7) before P4

main spot

(8) at T-shape beam stop

~ 5 mm

(9) at 1st amp C102

pinpoint (barely seen)

(10) at P6

(11) at Bethune cell

(12) at M17

brighter

M17
Optimization of the position of the cylindrical lenses
## Appendix A.2: Data for optical elements in the system

### Lenses

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<td>20</td>
</tr>
<tr>
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### Mirrors

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<td>J</td>
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**Dye cells**

**Apertures sizes (mm)**

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**Components**

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**Suppliers**

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<td>D-8215</td>
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<td>Tel # 161 13201</td>
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</table>

i) Normal dye cells

ii) Transmission grating 55 λ/mm fused silica with metal lines
iii) DFDL cell
   custom made by Max Planck Institute

iv) Bethune cell
   custom made by Max Planck Institute

v) Microscope objective
   Leitz Wetzlar

vi) BBO crystal, 52°,
    4 x 4 x 0.3 mm in fused
    silica cell; Index matching
    fluid - FC-43.
    Photox Optical Systems
    P.O.Box 274
    Huntington, Oxford
    OX 3035, U.K.

vii) SCDL cell
     LO Laseroptic GmbH
     D-3008 Gahisen 8

viii) Saturable Absorber
      Schott OG 530
      Schott Glass Inc.

ix) DC motor and Gear
   Fau haber
   Postfach 1146
   D-7036 Schönäich

x) Immersion oil for DFDL cell
   Zeiss Immersionsoel 518 C
   DIN 58 844 ISO 8036/1
   \( n_e = 1.518 \) (23°C)
   Zeiss
Appendix A.3: Data for all dyes in the system

### Dye Concentrations

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</table>

### Pumping Levels

- Quenched Cavity Dye Laser: $5 \times$ threshold (at least 3 times)
- Short Cavity Dye Laser: $5 \times$ threshold (at least 3 times)
- Gated Saturable Absorber: $5 \times$ threshold (at least 3 times)
- Distributed Feedback Dye Laser: $2 \times$ threshold
- Saturable Absorber: $5 \times$ threshold (at least 3 times)
### Appendix A.4: Excimer Laser Gas Mixtures

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<th>KrF laser</th>
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<td>F₂ (5% in He)</td>
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<tr>
<td><strong>Total</strong></td>
<td><strong>Total</strong></td>
</tr>
<tr>
<td>2600 mb</td>
<td>2500 mb</td>
</tr>
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</table>
Appendix A.5: Synchronization of the KrF amplified laser beam

The synchronization of the amplified KrF laser beam can be measured by monitoring the ASE background. Due to the saturated amplification of the second amplified beam, the KrF gain will be depleted and the depletion is observed within the gain recovery time. The timing of the first amplified KrF beam should be adjusted around the peak of the ASE background (KrF gain) and approximately 5 ns prior to the second amplified beam.

A biplanar vacuum photodiode (Hamamatsu) is used to monitor the timing of the KrF amplified beam with an aperture and ND filter(s) for beam attenuation. A typical temporal profile of the ASE with and without the seed beam is shown in the next page.

Figure A.5.1. Setup for the timing measurement
Figure A.5.2. Typical temporal profile of the KrF ASE background
Appendix A.6: 1-m spectrometer for analysis of laser spectra

A one meter Littrow-type spectrometer is built for the analysis of the spectra of the ultrashort laser radiation. A diode array is used to monitor the spectra of both the KrF laser radiation and the fundamental green radiation.

![Diagram of the spectrometer setup]

Figure A.6.1 Schematic diagram of the spectrometer for the spectral analysis
<table>
<thead>
<tr>
<th>Component</th>
<th>Description</th>
<th>Manufacturer</th>
<th>Model/Part Number</th>
</tr>
</thead>
<tbody>
<tr>
<td>Variable slit</td>
<td>slit width variable from 0 to 3 mm resolution 10 μm</td>
<td>Spindler-Hoyer</td>
<td>036105</td>
</tr>
<tr>
<td>CaF₂ lens</td>
<td>f = 100 cm, 2 inch diameter</td>
<td>Janos</td>
<td>A1404V575</td>
</tr>
<tr>
<td>Diffraction grating</td>
<td>3000 lines/mm 34 x 34 cm blaze wavelength = 250 nm aluminum coating</td>
<td>American Holographic</td>
<td>100.3000-P2-UV</td>
</tr>
<tr>
<td>Photodiode array</td>
<td>1024 diodes 25 μm centers, 25μm aperture</td>
<td>EG&amp;G</td>
<td>RL1024SAQ-011</td>
</tr>
<tr>
<td>Video board</td>
<td>sample and hold output maximum clock frequency = 250 kHz</td>
<td>EG&amp;G</td>
<td>RC1000/RC1001</td>
</tr>
<tr>
<td>Grating mounting</td>
<td>360° rotary stage</td>
<td>Microkinetics</td>
<td>CTCCP</td>
</tr>
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<td></td>
<td>tilting stage</td>
<td>Microkinetics</td>
<td>CTC57190</td>
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<tr>
<td></td>
<td>prism holder micro stage</td>
<td>Spindler-Hoyer</td>
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</tr>
</tbody>
</table>
Appendix A.7: Polarization of the ultrashort pulse excimer laser beam

It is important to know the degree of polarization of the ultrashort excimer laser beam for any applications of the laser, especially laser-target experiments. There are several ways to measure the polarization of the UV laser pulse, however, we apply a simple method to measure reflection of the laser beam with a fused silica plate under the Brewster angle.

The excimer laser beam is expected to have a polarization parallel to the laser table. Since the dye cells are side-pumped the green dye laser beam has a polarization perpendicular to the laser table, which is necessary to the frequency doubling at an appropriate crystal orientation. Just after the nonlinear crystal, the doubled UV laser beam should have a polarization direction perpendicular to the table. The direction of the polarization, however, is rotated 90° after the beam is bent by two mirrors in order to feed the beam to the excimer amplifier which is located beneath the dye laser system. Assuming the direction of the polarization parallel to the table (or ground), the degree of polarization has been measured using a fused silica plate under the Brewster angle. The degree of polarization was also determined by measuring the transmission of the laser through a Glan-Thompson polarizer as a function of the angle of rotation of the polarizer. The results of the measurement are shown in Figure A.7.1. These results agree with the polarization measurements made with a fused silica plate under the Brewster angle.
Degree of Polarization = $T_{\text{max}} - T_{\text{min}} / (T_{\text{max}} + T_{\text{min}}) = 76.5\%$
Appendix B

Photodiode array for the one meter spectrometer
The purpose of the photodiode array and its peripheral board is to convert the optical image of the laser spectra to a set of electrical signals. A schematic diagram for the photodiode array detection circuit can be found in Figure 2.1. The photodiode array itself [Reticon model S series # RL1024SAQ-011] is one inch long and is composed of 1024 photodiodes spaced 25 μm apart; the width of each diode is 25 μm. Light incident on each diode is converted to electrical charge that is stored on the diode capacitance until readout. Readout is initiated by a START signal and controlled by a clocked shift register. The process of scanning the photodiode array is handled by the video peripheral board [Reticon board # RC1000/100]. A modified trigger signal from the excimer laser (5 volt, 10 μs square pulse) is used to synchronize the scanning of the photodiode array with the arrival of the laser pulse at the photodiode array. Fed into the external START port of the diode array board, the trigger signal causes the video board to begin scanning the array. The final sampled and held video output signal can be monitored from the VIDEO OUT port. Since the clock frequency of the video board is set at 100 kHz, one full scan of the 1024 diodes in the array can be viewed when the oscilloscope time scale is set a 1 mv/div.

Detailed information on the operation of the diode array or alignment of the video processing board can be found in the RC1000/1001 manual from EG&G Reticon. It should also be noted that the diode array is extremely sensitive. This usually means that all room lights should be turned off while the diode array is running. As a general guideline, the video output signal should be less than 2 volts during normal operation. The video output becomes saturated around 6 volts.

Also, it has been observed that the clock frequency of the video board tends to drift to a slower frequency while the diode array is on. The effect is minimal over the span of ~ 3 minutes. However, if the diode array is on for ~ 10 minutes, the width of the array on oscilloscope screen will be noticeably wider than when the array was first activated. This
effect should be kept in mind when making any type of quantitative analysis of the laser spectra captured by the diode array.