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Development and Applications of A High Intensity Ultrashort Pulse UV Laser System

by

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Blackett Laboratory Laser Consortium -- High Intensity Ultrashort Pulse UV Laser System

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ABSTRACT

This programme of research incorporates work on the development of a high intensity ultrashort pulse UV laser system for investigations on the interaction of intense laser radiation with matter, such as multiphoton ionisation of atoms at high intensities as well as linear and nonlinear atomic and molecular spectroscopy.

The high intensity laser system starts with the generation of ultrashort visible seed oscillator pulses, which are then amplified in a multistage dye amplifier chain to mega-watt power level, frequency doubled to the UV spectral region, and then boost up in a KrF amplifier. The visible source has a wide range of wavelength tunability. Both the visible and the UV source could be operated at manual single shot mode or up to tens and hundred of hertz repetition rate.

The wavelength stability of the visible blue-green seed oscillator has been optimised and further characterised to meet the maximum bandwidth gain of the KrF amplifier. We can not only boost the seed oscillator pulses in the dye amplifier chain with best amplification and maximum energy extraction but can also self-select a single amplified pulse by the optimisation of the interaction volume of the pump beam and the seed beam. We have optimised the operating conditions of the KrF UV amplifier at the edge of the saturation region to obtain efficient amplification and better output shot to shot stability.

A high focused intensity greater than 10^{16} W/cm^2 , ultrashort pulse of 200 fem-

toseconds duration, ultraviolet 248.6 nm laser system has been developed. It could be further improved to much higher intensities by reducing the seed oscillator pulse duration and by using pulse compression techniques to minimise the pulse broadening and compress it to the shortest amplified pulse. This system consists of several useful sub-systems for specific applications. Furthermore, since we have mastered the techniques of developing high intensity laser systems, we can use this system as a front end, and cascade further, amplifying this system output to the extremely high intensity of over $10^{20} W/cm^2$.

With this high intensity source we can provide a significant contribution to a new and exciting field of fundamental Physics.

Chapter 1

Introduction

1.1 Motivation

A programme of research on the interaction of intense laser radiation with matter incorporating work on the development of high intensity ultrashort pulse laser systems, multiphoton ionisation of atoms at high intensities as well as linear and nonlinear atomic and molecular spectroscopy is underway in the Blackett Laboratory Laser Consortium.

One important emphasis is the study in which pulses of radiation with very high intensities interact with atoms. By amplifying ultrashort pulses of sub-picosecond duration to even modest energies of .01 to 0.1 Joule, it is possible to create intensities of 10^{16} to 10^{17} W/cm^2 by focusing with a simple optical system. At these high intensities, the optical electric field becomes comparable to or greater than the Coulomb field in an atom. The theoretical models involving lowest order perturbation theory that are generally used to describe the interaction of intense laser radiation with matter are no longer valid. At high intensities, multiphoton ionisation is observed, although many of the experiments carried out so far may still be interpreted in terms of sequential single step processes, there is evidence of coherent coupling of several electrons to the radiation field giving rise to multiply excited high lying states. The higher intensity laser systems currently being developed should ensure more definitive experiments to

be carried out and permit a more accurate picture of the basic physics involved in these interactions.

6 photon ionisation of Xenon by a picosecond Nd:YAG laser pulse was first observed in 1979^[1.1]. With now almost routine intensities of around $10^{12} W/cm^2$, it becomes possible to drive continuum-continuum transitions^[1.2]. This enables us to observe above threshold ionisation (ATI) in the electron spectrum. At intensities around $10^{13} W/cm^2$ the kinetic energy due to the electron wiggle in the laser field becomes comparable to the laser photon energy. This ponderomotive potential influences the photoelectron spectra dramatically, shifting and deleting the peak produced^[1.3]. In the paper by Woerkom et al^[1.4], the role of Stark shift induced resonances is discussed. The article shows how the spatial and temporal variation of the electron spectra produced depends on an interplay between the bound state shifts and widths with dressed electron wavepacket escape. The variation of the electron spectra with pulse duration, pulse shape and intensity has proved to be a superb probe of ATI dynamics.

Knight and Shore^[1.5] using a theory of coupling to electron continua dressed by continuum-continuum transitions predict the production of high order harmonics along with ATI electron peaks. This raised the possibility of direct production of coherent XUV by very high order processes. The subsequent experimental observation by Ferray et al^[1.6] along with the related work of McPherson et al^[1.7] has provoked a concerted attack on the detailed description of the evolution of an electron in a field of around $10^{14} W/cm^2$. Shore and Kulander^[1.8] have also examined the effect of phase

matching on the up conversion process as this can enhance the efficiency. Eberly has an excellent review article^[1.9].

Recent developments in laser technology and in atomic and quantum optical theory have only just started to have an impact on what physicists perceive to be possible in multiphoton physics of atoms in an intense field.

In the past few years, techniques have become available which enable laboratory scale lasers with output powers of terawatts to be set up. The key to these developments is the generation and amplification of ultrashort pulses with sub-picosecond duration^[1.10]. A terawatt laser of 1 ps pulse duration and 1 J output has been developed by Mourou et al ^{[1.11][1.12]}. This laser can be accommodated on an optical table and gives rise to the title of "table top terawatt" (T^3) laser.

The significance of this high intensity can be seen from the comparison of the electric field of the laser radiation and the Coulomb field in an atom. For the hydrogen atom the Coulomb field at a Bohr radius a_0 from the nucleus is

$$E = \frac{e}{4\pi\epsilon_0 a_0^2} = 5.14 \times 10^9 V/cm \quad (1.1)$$

The relationship between the laser intensity I and the electric field E is given by $I = \frac{1}{2}\epsilon_0 c E^2$, where ϵ_0 is the permittivity of the vacuum and c is the speed of light. Or simply as

$$E[V/cm] = 27.4 I^{\frac{1}{2}} [(W/cm^2)^{\frac{1}{2}}] \quad (1.2)$$

Thus for a terawatt laser at $1\ \mu\text{m}$ wavelength which can be focused to an intensity of $\sim 10^{19}\text{W}/\text{cm}^2$, an electric field of about $9 \times 10^{10}\text{V}/\text{cm}$ will be generated. If the atom can survive the rapidly increasing intensity of the ultrashort pulse from a terawatt laser the electrons may be exposed to electric fields higher than that due to the nucleus.

1.2 Laser development

To characterise a laser system that delivers intense laser radiation, there are two important linked parameters: (1) the pulse duration, and (2) the output energy or power. Intense laser radiation sources could be achieved either by extremely high output energy laser systems or by the generation of ultrashort pulses at moderate output energy levels that give high output power and can consequently be focused to very high intensity.

Short pulse (nanosecond and picosecond) and ultrashort pulse (a few picosecond and femtosecond) generations have progressed remarkably in the past three decades. The ability to generate short pulses has advanced by about 8 orders of magnitude over this period. Indeed, sub-hundred femtosecond pulses can be obtained routinely nowadays in the laboratory or even in commercial systems. This is the easiest way to obtain a high intensity laser system.

To construct a high intensity laboratory scale laser system we must consider first how to generate the ultrashort seed pulse, and what the power level of this seed

pulse is; and secondly what kind of amplifier stage(s) can boost the ultrashort pulse output power to the desired range together with the necessary frequency conversion for the final power amplifier stage, and still preserve the good beam qualities.

Dye lasers can generate ultrashort pulses of sub-picosecond pulse durations and less. Direct generation of 27 femtoseconds pulses in a colliding pulse mode-locked ring dye laser has been reported^[1.13]. Recently 50 femtosecond pulses generated from the Ti: sapphire laser^[1.14] give another prospect for the generation of ultrashort pulses.

For the gain amplifier there are three important requirements : (1) a sufficiently large bandwidth to amplify the short pulses, (2) the efficient energy extraction of the laser radiation from the amplifier, and (3) the preservation of the good beam quality to produce high intensity.

Typically, dye and excimer systems have broad bandwidths of 20 - 40 nm which can support pulses as short as 30 fs. Solid state laser materials have similar or even greater (exceptionally as large as of 200 nm for Ti:sapphire) bandwidths. However the principal difference between solid state materials and dye or excimer systems is their saturation conditions that determine the efficiency of amplifiers. The saturation energy density E_s is $h\nu/\sigma$ with unit J/cm^2 , where ν is the optical frequency and σ is the stimulated emission cross section. For dyes and excimers, the stimulated emission cross sections are relatively high, $\sigma \sim 10^{-16} cm^2$. This leads to a relatively low saturation energy density of a few mJ/cm^2 . Solid state materials have much smaller cross sections of $\sigma \sim 10^{-20} cm^2$, leading to very large saturation energy densities of up to 5 J/cm^2 . But in the solid state laser systems, if a pulse with the minimum

pulse duration t_p allowed by the gain bandwidth is passed through an amplifier at the maximum energy density for efficient energy extraction, the resulting power density E_s/t_p of $\sim 10^{14} W/cm^2$ is well above the critical intensity of catastrophic damage to the laser materials.

High intensity pulse will produce severe distortion of the beam quality by the nonlinear refractive index in the amplifiers. The change in refractive index induced by a pulse of intensity I is given by

$$n = n_0 + n_2 \langle E^2 \rangle = n_0 + \gamma I \quad (1.3)$$

where γ is a coefficient which depends on the wavelength and material. This effect may be quantified from the additional phase change produced by the beam along a medium of length L given by^[1.15]

$$B = \frac{2\pi\gamma}{\lambda} \int_0^L I(z) dz \quad (1.4)$$

A critical intensity at which the beam is significantly distorted occurs when $B \sim 5$, which corresponds to an intensity of $\sim 10^{10} W/cm^2$. While this intensity is comfortably above that corresponding to gain saturation for a dye or excimer medium of $I_s = E_s/t_p = [2mJ/cm^2]/[1ps] \sim 10^9 W/cm^2$, it is very much less than the corresponding figure of $\sim 10^{14} W/cm^2$ for a solid state amplifier. It follows that while dyes and excimers are very satisfactory amplifiers for sub-picosecond pulses of a few millijoules energy, the potential of solid state media as efficient amplifiers cannot be

realised due to nonlinear effects. However this disadvantage can be overcome by the chirped pulse amplification technique^[1.12].

The saturation energy density of a few millijoules certainly limits the maximum output energy obtained in dye and excimer amplifiers. These media can only be used for amplifying pulses of high energy if the area of the aperture of the amplifier is large. The aperture of dye amplifiers is generally employed to the maximum of a few millimeters because of the limited penetration depth of the pump radiation. Excimer lasers^[1.16], however, are gaseous systems and being pumped by electric discharges or high current electron beams do not have the same disadvantages as the pumping of dye amplifiers. With either UV radiation or x-ray pre-ionisation, stable discharge apertures of up to 5 to 10 cm may be produced. Larger apertures up to 50 cm or more may be pumped using a high current pulsed E-beam.

The ultrashort seed pulses after converting their wavelengths to match the gain bandwidth of the rare gas halide excimers can be amplified in successive excimer amplifiers of increasing apertures by use of beam expanding optics to maintain the energy density of the beam close to the saturation value E_s for best amplification. In the excimer system at Rutherford Appleton Laboratory pulses of 3.5 ps duration are amplified in a series of discharge pumped and E-beam pumped amplifiers of increasing apertures to an energy of 2.5 J^[1.17]. This is the highest energy so far obtained in an ultrashort pulse.

Table 1.1 summaries the properties of various amplifier materials for comparison.

**Table 1.1 Comparison of relevant properties
of various amplifier materials**

Medium	Wavelength (nm)	Gain Bandwidth (nm)	Pulsewidth (fs)	Saturation Energy (J / cm ²)
Dye	450 -- 850	30	30	1.6×10^{-3}
Excimer (KrF)	248	5 -- 10	50	2×10^{-3}
Nd : Glass	1064	20	300	5
Ti : Sapphire	800	200	10	9×10^{-1}

Medium	Stimulated emission cross Section (cm ²)	Saturation intensity (GW / cm ²)	Focused intensity (W / cm ²)	B (L = 10 cm)
Dye	2×10^{-16}	50	10^{19}	< 4
Excimer (KrF)	1×10^{-16}	40	10^{19}	< 4
Nd : Glass	4×10^{-20}	5×10^3	5×10^{21}	2000
Ti : Sapphire	2.8×10^{-19}	9×10^3	7×10^{20}	2000

1.3 High intensity ultrashort pulse UV laser system

Our laboratory scale high intensity ultrashort pulse UV laser system is produced following the steps of the architecture in Figure 1.1.

The first step is ultrashort pulse generation at a visible wavelength. It has two important functions: (1) it alleviates the difficulties of directly producing a very short pulse in the UV region; (2) the laser noise can be reduced sharply by introducing an intermediate nonlinear process. There are a number of dye lasers that produce pulses with durations from a few hundred femtoseconds down to a few tens of femtosecond. And there is a wide range of active/passive dye combinations that can generate ultrashort pulses from far UV to near IR wavelengths as shown in Figure 1.2.

It is very important to select a pair of active/passive dye combination that can tune the laser pulses to the desired wavelengths to match the excimer lines. There has been a proposal that by using C102/DOCI dye combination^[1,18], sub-hundred femtosecond pulses can be generated in the Blue-Green wavelength region tunable to 497.2 nm that is exactly the double wavelength of the KrF amplifier at 248.6 nm. It has been realised in this development that indeed the C102/DOCI CPM ring dye laser with an intracavity dispersion compensating prism sequence can generate ultrashort pulses of 88 fs, with output power 10 mW at 497.2 nm.

The ultrashort seed oscillator pulses from the C102/DOCI CPM ring dye laser were amplified in a chain of dye amplifiers, frequency doubled to 248.6 nm and amplified in the KrF amplifier. The results were not completely satisfactory and we have found that the wavelength of the seed oscillator was jittering. Because we need to

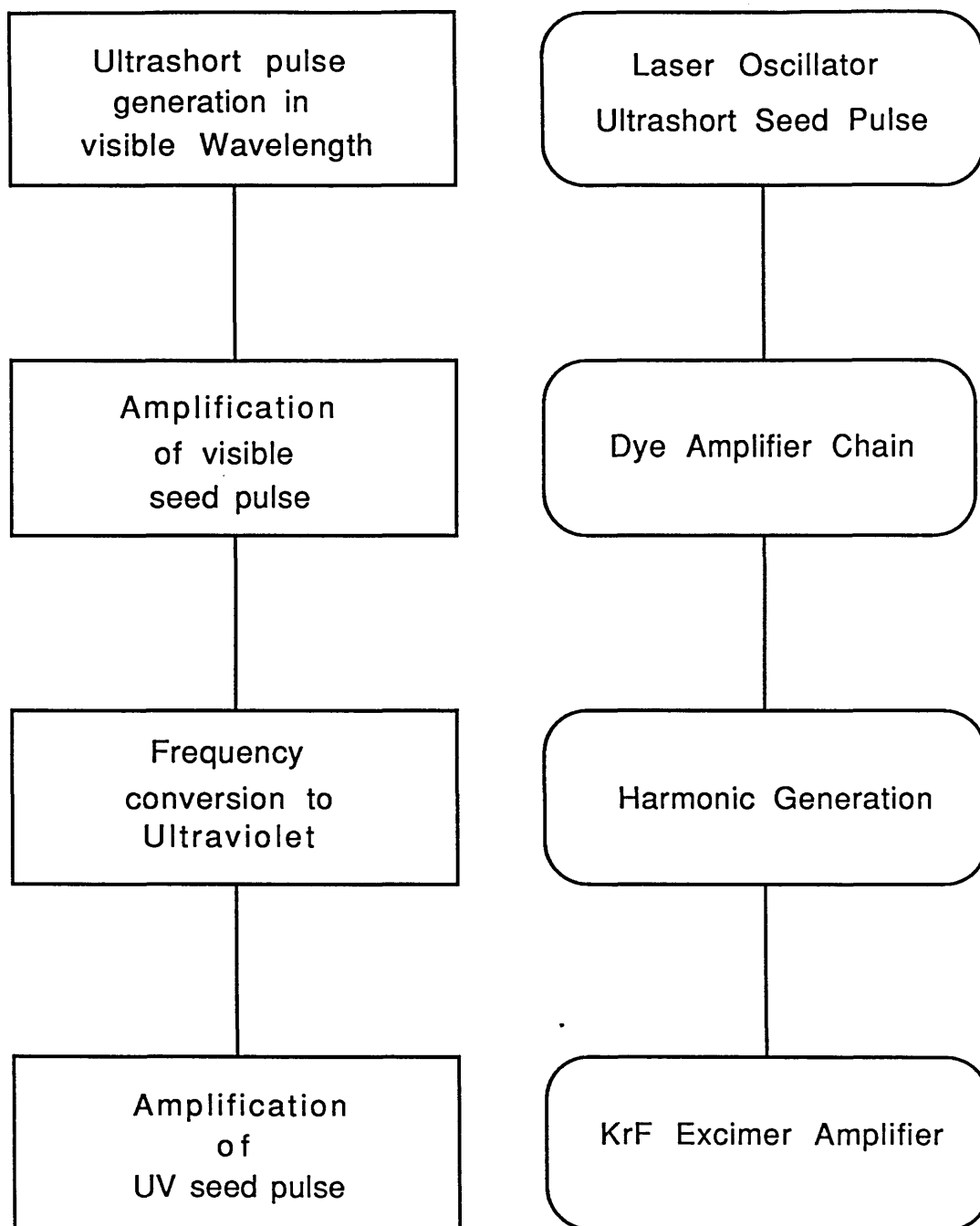


Figure 1.1 Architecture of building up high intensity ultrashort pulse UV laser system and our system block diagram.

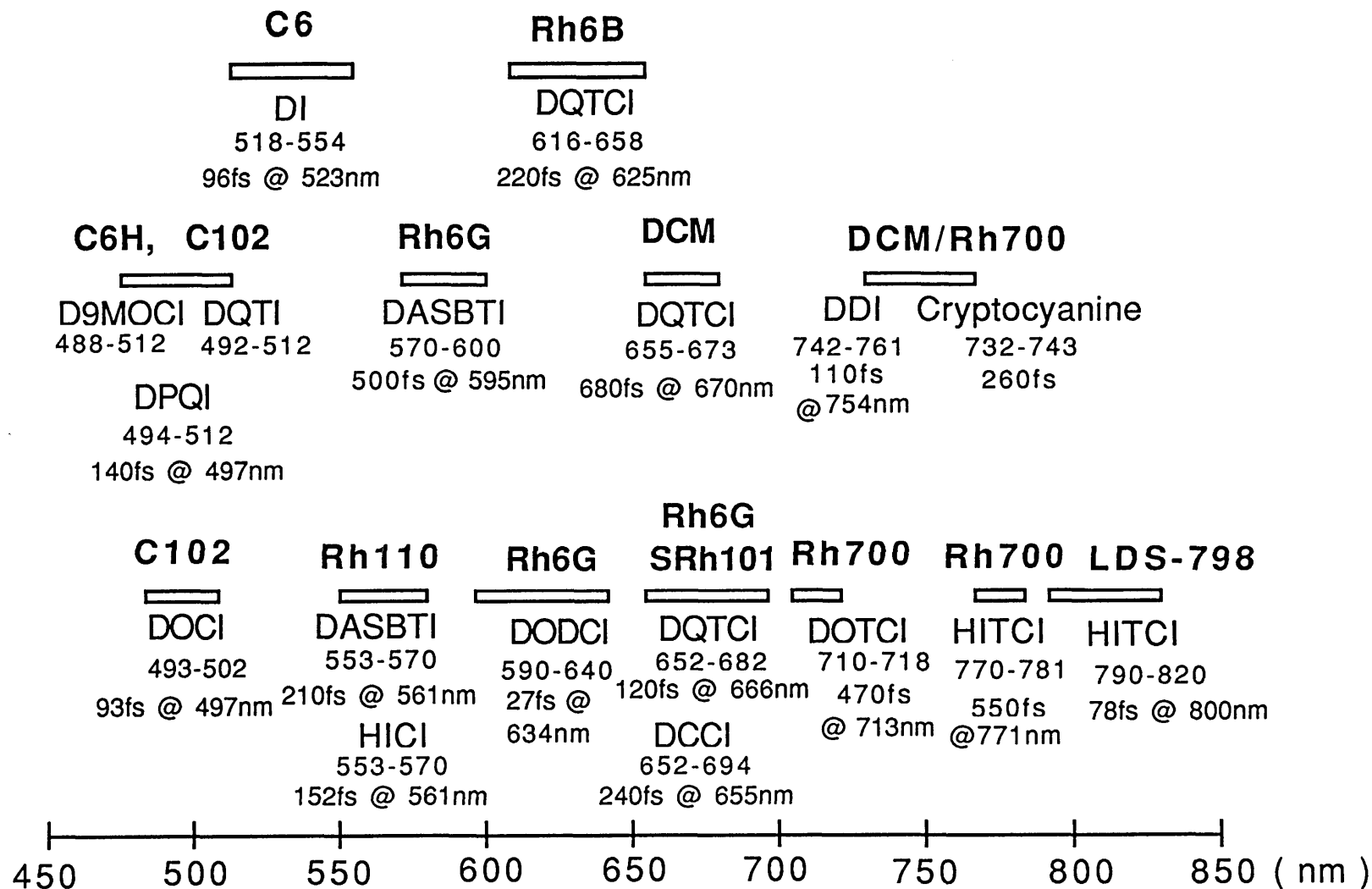


Figure 1.2 Currently available tuning ranges of passively mode-locked CW dye lasers

set up a stable system for the experimental applications within at least a reasonable time period, we needed to optimise the wavelength stability and characterise the wavelength jittering if it is inevitable.

Amplification of the seed oscillator pulses is necessary to produce a strong seed beam. The pulses emitted by femtosecond laser oscillators are frequently not sufficiently energetic for a particular application, and amplification is necessary. For instance, peak powers delivered by passive mode-locked systems are typically $\sim KW$, whereas powers in the MW and GW range are often required. Many of the basic considerations in pulse amplification are applicable to dye amplifiers. A good amplifier should minimise distortion in the temporal, spatial and spectral properties of the signal pulse. Since the amplifier dye is a high gain medium the suppression and elimination of the amplified spontaneous emission (ASE) is most important in this second stage of the system architecture. Both spatial filter and the Schott colour glass filter saturable absorber were employed and have quite effectively suppressed the ASE. Temporal broadening can also be caused by group velocity dispersion from the dye solution and optical materials like windows and lenses. This effect rapidly becomes significant when the pulse duration becomes shorter, because it scales inversely to the square of the pulse duration^[1,19]. Spatial distortion from amplifiers will rapidly degrade the ability to focus to high intensity. Distortions often result from the gain region not being pumped uniformly, thermal turbulence in the fluid flow, and self focusing effects in the gain medium. Transversely pumped dye cuvettes are found to be satisfactory for small beam amplification. It is difficult to produce uniform amplification for beam diameters larger than a few millimeters without compromising

efficiency severely. In this development we have found out that by optimally aligning the interaction volume of the seed beam and the pump beam we can suppress the ASE, obtain best amplification, maximum energy extraction and achieve single amplified seed pulses.

Frequency conversion is achieved with the nonlinear crystal Beta Barium Borate ($\beta - B_aB_2O_4, BBO$). The main concern of this process is the thickness of the crystal. Crystal thickness must provide sufficient phase matching bandwidth for the ultrashort pulse, and the amount of pulse stretching due to group velocity mismatch of harmonic and fundamental waves must be tolerable. These requirements drastically limit conversion efficiency because it scales as the square of the crystal length.

An excimer laser has properties that are most suitable for a high power amplifier system. It exhibits the properties of wide gain bandwidth for ultrashort pulse amplification, high gain, large energy extraction efficiency, high self-focusing threshold and scalability to large apertures. In particular, a KrF excimer amplifier, which has the highest gain, has a photon energy of approximately 5 eV. The laser transition of the KrF^* excimer $B^2\Sigma(1/2) \rightarrow X^2\Sigma(1/2)$ is bound free in nature, which produces a broad essentially structureless emission spectrum. The linewidth of the transition is approximately 320 cm^{-1} as determined by the Franck-Condon overlap of the $v = 0$ excited state and the repulsive ground state, a value which is sufficient to amplify pulse as short as 17 fs, since $(2\pi\delta\nu)^{-1} = \gamma_1^{-1} \sim 17 \text{ fs}^{[1,20]}$.

Despite its large homogeneous gain bandwidth, the emission cross section is approximately $2 \times 10^{-16} \text{ cm}^2$, a value that is much larger than most solid state gain

media. The short fluorescence lifetime of 6.8 ns contributes to this cross section. In this case, the short lifetime is matched to the typical time scale of pulse discharge which is normally tens of nanoseconds. Standard commercial units can exhibit a small signal gain and length product of 9.9, and the energy extraction efficiency for ultra-short pulses of less than 5 ps can be as high as 75 percent^[1,21]. The saturation energy of the KrF amplifier is approximately 2.5 times higher than the XeCl amplifier^[1,21]. And for the output energy consideration, it can be scaled to large aperture.

A potential limitation to the power is two photon absorption in the window material. However high purity calcium-fluoride CaF_2 and magnesium-fluoride MgF_2 materials have proven to be satisfactory materials for power densities up to $50\text{GW}/\text{cm}^2$ ^[1,22].

After the first amplification in the KrF amplifier, the beam is spatially filtered by a $50\text{ }\mu\text{m}$ vacuum spatial filter to suppress the KrF amplifier ASE. We have double passed the beam in the same KrF amplifier as a final amplification.

Figure 1.3 shows the complete schematic diagram of our high intensity ultra-short pulse UV laser system.

We have used the C102/DOCI active and passive dye combination to directly generate ultrashort pulses of 200 fs pulse duration in 497.2 nm that can be frequency doubled for the KrF final amplification. The seed pulses were first amplified in the dye amplifier chain to a much higher output power for the second harmonic conversion. The dye amplifier chain can amplify the seed pulse with best amplification, maximum energy extraction and a self-selected single amplified pulse.

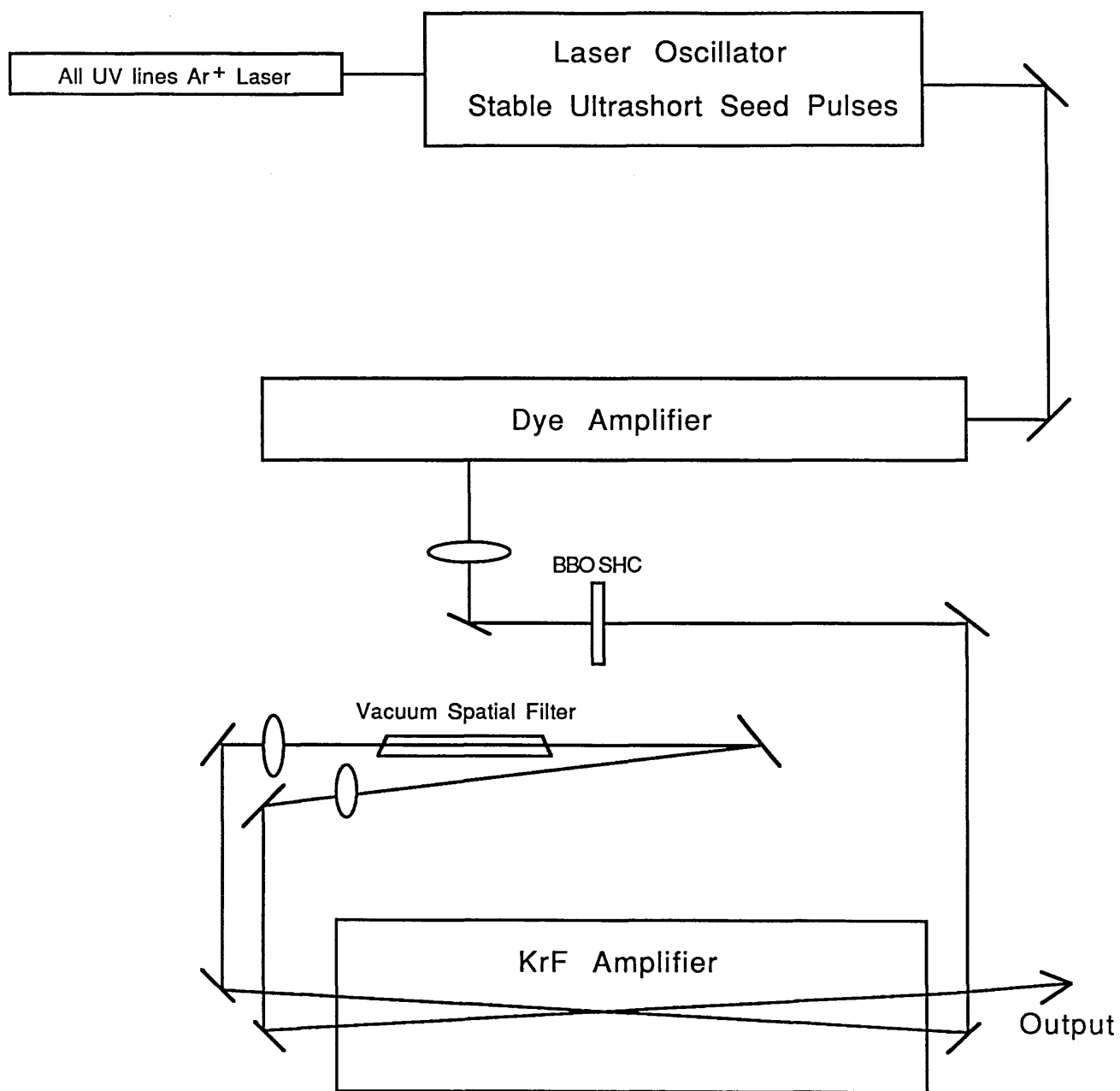


Fig. 1.3 High intensity ultrashort pulse UV laser system.

The second harmonic crystal BBO can efficiently convert the visible seed pulse to a UV pulse which is double pass amplified in the powerful KrF amplifier. It is an efficient and self-contained high intensity ultrashort pulse UV laser system capable of generating peak powers of 6 GW and with peak intensities of 10^{16} to 10^{17} W/cm^2 . What is more, after the dye amplifier stage the system could be employed as a tunable visible sub-system between 492 nm and 503 nm.

The stable seed oscillator section consists of a linear cavity dispersion compensated passively mode-locked C102/DOCI dye laser, the dye laser pump source of the all UV lines Ar^+ ion laser, and the diagnostic set up. The diagnostic set up includes a fast response avalanche photodiode that displays the mode-locked pulse train on the oscilloscope and provides the trigger source for the system synchronisation, a real time autocorrelator that can optimise the generation of the shortest ultrashort pulses, a spectrum meter and a wavemeter that monitor the seed beam wavelength stability.

Table 1.2 lists the relative high intensity ultrashort pulse visible and UV laser systems for comparison. Our approach is an efficient and simple one to achieve the goal, it could also provide various subsystems for different applications and act as the front end for the ultimate extreme high intensity system.

Table 1.2 High intensity ultrashort pulse laser system

Amplifier	Seed Oscillator Source	Output		Institute	Reference
		Pulsewidth	Energy		
XeCl (EMG101-MS)	Colliding pulse mode-locked dye laser, 616 nm, 4 stages dye amplifier chain, 2w (KDP),	160 fs	12 mJ	IBM T. J. Watson Research Centre	JOSA B, 4, 1061, 1987
XeCl (Wide aperture 7cm x 7cm)	Synchronously pumped dye laser (R590), 616 nm, 4 stages dye amplifier chain, 2w (BBO),	310 fs	300 mJ	Institute for solid state Physics, Univ. of Tokyo	Optics Lett. Vol 13, 580 1988
Dye Amplifier (4 Stages 5 passes)	Synchronously pumped dye laser, 620 nm, Colliding pulse mode-locked dye laser (CPM), CPM and Pulse compression	900 fs	1 mJ	Laval Univ. & Div. of Physics National Research Council of Canada,	Optics Lett. Vol 59, 64, 1986 Phy. Rev. Lett. Vol61, 153, 1988
		90 fs	550 uJ		
		22 fs	110 uJ		
Dye Amplifier (5 stages 6 passes)	Colliding Pulse mode-locked dye laser (R6G/DODCI),	106 fs	2.5 mJ	Dept. of Phy. University of Berkeley	JOSA B, Vol 5 1573, 1988
Dye Amplifier (4 stages 5 passes)	Synchronously pumped dye laser (R6G), 570-600 nm,	1 - 2 ps	3 mJ	Dept. of Nuclear Engineering, Univ. of Berkeley & LLNL	Phy. Rev. A , 37, 747, 1988 Optics Lett. 14, 42, 1989

Amplifier	Seed Oscillator Source	Output		Institute	Reference
		Pulsewidth	Energy		
KrF (EMG-150) (single stage, double passes)	Distributed feedback dye laser (DFDL) 497 nm, 2 stages dye amplifier chain, 2w (BBO),	80 fs	15 mJ	Max-Plank Institute	O. C. 63, 305, 1987 O. C. 68, 186, 1988 App. Phys. B, 239 1989
		60 fs	13 mJ		
		150 fs	45 mJ		
KrF (EMG 201 MSC) Prometheus amplifier (large aperture KrF amplifier)	Hybrid mode-locked dye laser 745 nm, 2 stages dye amplifier chain, Frequency doubling & mixing, 745 nm -- 372 nm -- 248 nm (KDP)	600 fs	250 mJ	Dept. of Phy. Univ. of Illinois	Optics Lett. 14,1113, 1989
KrF (EMG 201) KrF (EMG 200) Two stages KrF amplifier	Synchronously pumped hybrid mode-locked dye laser, 648 nm 3 stages dye amplifier chain, Frequency doubled and sum- frequency mixed 648 nm -- 324 nm -- 248 nm (BBO)	700 fs	25 mJ	Los Alamos National Laboratory	Opt. Lett. 13, 734, 1988
Two KrF amplifier Double pass large aperture KrF amplifier (7cm x 7cm) Double pass e-beam-pump KrF amplifier) (23cm x 23cm)	Synchronously pumped dye laser, 746 nm, 4 stages dye amplifier chain, Frequency tripled in 1 mm KDP and BBO,	390 fs 280 fs	1.5 J 410 mJ	Institute for Solid state Physics, Univ. of Tokyo	Opt. lett. 14, 353, 1989 Opt. lett. 15, 845, 1990

Amplifier	Seed Oscillator Source	Output		Institute	Reference
		Pulsewidth	Energy		
KrF (EMG103 MSC) discharge pumped pre-amplifier	Synchronously pumped dye laser, 746 nm, 3 stages dye amplifier chain, Frequency tripling by 2 KDP, 746 nm -- 372 nm -- 248 nm	3.5 ps	100 uJ	Laser Division Rutherford Appleton Laboratory	Opt. Comm. 66,127, 1988
Double pass GOBLIN e-beam pumped KrF pre-amplifier (dia= 60 mm)		3.5 ps	20 mJ		
Double pass SPRITE e-beam pumped KrF amplifier (dia= 270 mm)		3.5 ps	2.5 J		
Double pass KrF (EMG201 MSC) discharge pumped amplifier (G= 7 E 4) (dia = 8 mm)	Colliding pulse mode-locked ring dye laser (CPM), C102/DOCl, 497 nm,	88 fs		Blackett Laboratory Laser Consortium Imperial College	
	Stable seed oscillator	360 fs 200 fs	25 pJ		
	3 stages dye amplifier chain (G=10 E 6)		50 uJ		
	2W (BBO), (eff. = 1%),		0.5 uJ		
			1.7 mJ		

1.4 The organisation of the thesis

This thesis consists of (1) the investigation and development of the stable seed oscillator source, (2) the optimisation of the dye amplifier chain for the best amplification, maximum energy extraction and the optimisation of a self-selected single amplified pulse in the visible region, (3) the second harmonic conversion and the amplification of the KrF amplifier, and (4) the immediate system applications for the investigation of above threshold ionisation (ATI) and high order harmonic generations.

We have investigated the generation of ultrashort pulses in the blue-green regime, and have achieved the shortest pulse duration of 88 fs at 497.2 nm at output powers of 10 mW. Detailed optimisation are stated in chapter 3.

Chapter 4 describes the further efforts of improving the ultrashort seed oscillator pulses wavelength stabilities and the quantitative characterisations of the wavelength stability relevant to the KrF gain bandwidth. The satisfactory stable seed oscillator source is then ready for the amplification stage through the multistage dye amplifier chain.

The dye amplifier chain is employed to preliminarily boost the seed pulse to a much higher output power. Optimisations of the dye amplifier chain for the best amplification, maximum energy extraction and the optimisation of the self-selected single amplified pulse in the visible region are the contents of Chapter 5.

Second harmonic generation of the seed pulse to the UV wavelength and the

amplification through the KrF amplifier are described in chapter 6. Chapter 7 concludes the development of this high intensity ultrashort pulse UV laser system and investigates the applications of the system to above threshold ionisation and high order harmonic generations.

In this research we have investigated and have a better understanding of the the generation of ultrashort pulses and have been able to obtain a stable ultrashort seed pulses as the source of the high intensity UV laser system. We can optimise the amplification of a self-selected single amplified pulse with best amplification and maximum energy extraction. We have also verified the amplification of the seed UV source in the KrF amplifier and have completely developed the high intensity ultrashort pulse UV laser system.

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Chapter 2

Ultrashort Pulse Generation and Measurement

2.1 Methods of generation

The time chronogram of progress in the generation of brief time intervals^[2.1] fall sharply in 1960 following the first operation of the conventional laser by Maiman in 1960^[2.2]. Since then, the technique of Q-switching by McClung and Hellwarth in 1962^[2.3] soon enabled giant pulse in the 10 - 50 ns regime to be produced. With the help of opto-electronic switching and chopping arrangements pulses of several hundred ps to a few ns duration can be obtained. The generation of shorter pulses in the picosecond regime (and further down into femtosecond regime) were first produced by passive mode-locking of a giant pulse ruby laser by Mocker and Collins in 1965^[2.4], and the Nd-glass lasers by DeMaria et al in 1966^[2.5]. Figure 2.1 shows the progress in ultrashort pulses generation since the invention of lasers in 1960. Up to now, dye lasers still dominate the field of direct generation of ultrashort pulses, and with extra grating pairs and prism pairs for high order dispersion compensation shortest pulse duration to date of 6 fs has been generated from a dye laser^[2.6]. Yet the Ti:sapphire active material has a much broader bandwidth than the dye and has been able to generate 50 fs pulses^[2.7]. In the very near future, this promising solid-state laser may be able to generate ultrashort pulses with durations as short as those produced

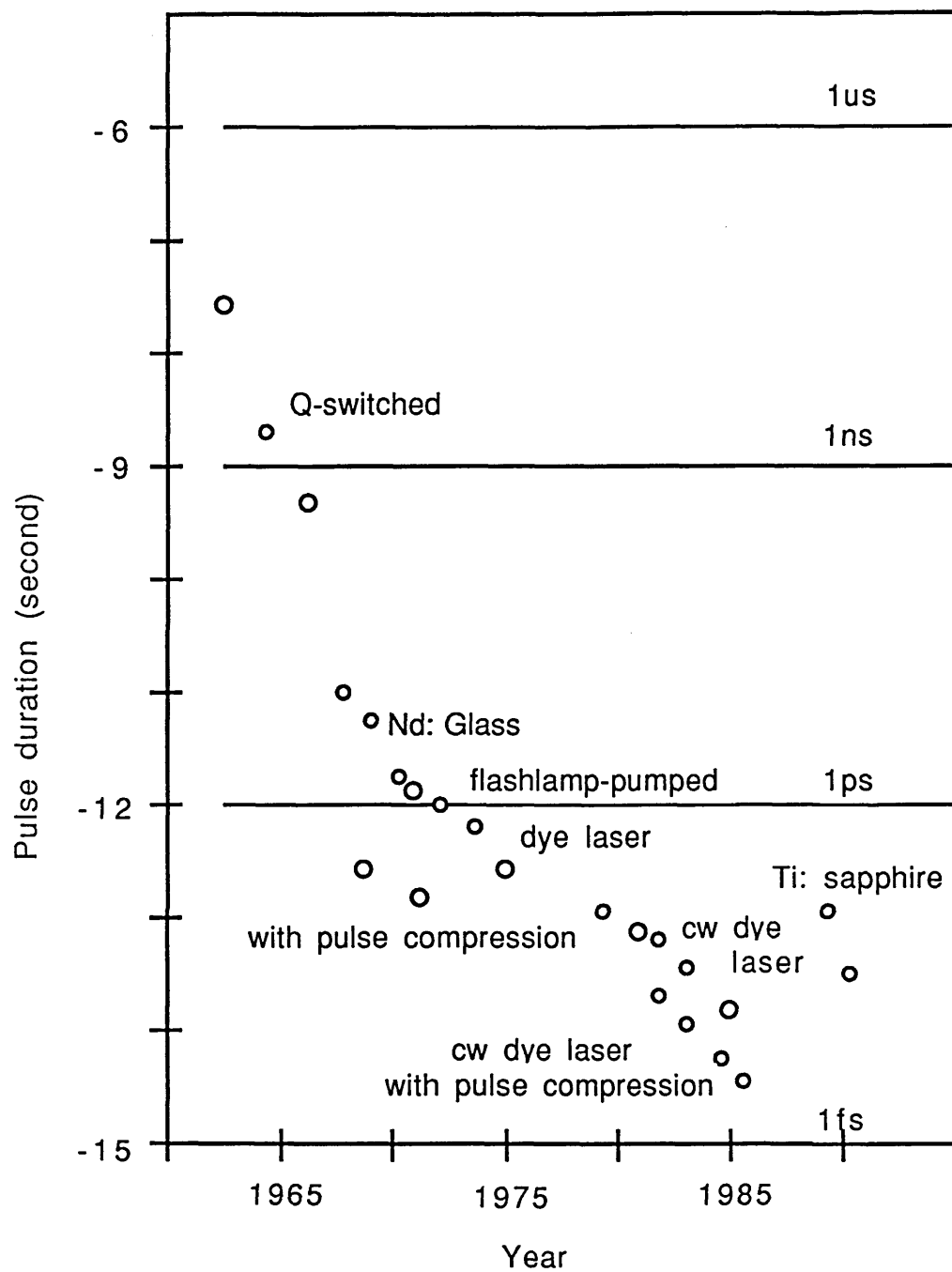


Figure 2. 1 Progress in ultrashort pulse generation since the invention of the laser in 1960. [similar to the figure that appears in G H C New, "Femtofacination", Physics World, Vol 3, P33, 1990.

with dye lasers. Typical examples of conventional lasers operating in the microsecond (μs) regime, Q-switched lasers in the nanosecond (ns) regime, and mode-locked lasers in the picosecond (ps) and femtosecond (fs) regime can now be found in many laboratories.^[2,8]

2.1.1 The principles of mode-locking

In conventional lasing conditions, the optical energy is spread out uniformly in the laser cavity, whereas in a mode-locked laser the energy is concentrated into a limited region of the cavity so that laser action involves a short pulse of optical radiation bouncing back and forth between the mirrors. For a linear cavity of length L , the pulse takes $2L/c$ for one cycle of the cavity if we neglect the refractive index effects. This is called the cavity transit time T_{cav} , that is $T_{cav} = \frac{2L}{c}$. Once per cycle, the pulse encounters the output mirrors of the cavity and a replica of the pulse is transmitted. The output of the mode-locked laser therefore consists of a regular sequence of pulses with periodicity T_{cav} and this is known as a mode-locked pulse train.

In an inhomogeneously broadened laser, oscillation can take place at a number of frequencies, which are separated by

$$\omega_q - \omega_{q-1} = \frac{\pi c}{L} \equiv \omega$$

The total optical electric field resulting from such multimode oscillation in the optical resonator can be taken as

$$e(t) = \sum E_n e^{i[(\omega_o + n\omega)t + \phi_n]} \quad (2.1)$$

where the summation is extended over all the oscillating modes, ω_o as a reference frequency, and ϕ_n is the phase of the n th mode. $e(t)$ is periodic in

$$T \equiv \frac{2\pi}{\omega} = \frac{2L}{c}$$

which is the round trip transit time inside the resonator.

Note that the periodic property of $e(t)$ depends on the fact that the phases ϕ_n are fixed. In typical lasers the phases ϕ_n vary randomly with time and this causes the intensity of the output to fluctuate randomly [2.9]. The technique of "mode-locking" is to force the mode phases ϕ_n to maintain their relative values that cause the oscillation intensity to consist of a periodic train with a period of $T \equiv \frac{2\pi}{\omega} = \frac{2L}{c}$.

To simplify the analysis take N oscillating modes with equal amplitudes $E_n = 1$, and assume $\phi_n = 0$. (Typically, E_n will have a Gaussian distribution of spectral amplitudes and random spectral phases.) [2.9] Then $e(t)$ gives

$$e(t) = \sum e^{i[(\omega_o + n\omega)t]} \quad (2.2)$$

with the summation taken from $\frac{-(N-1)}{2}$ to $\frac{(N-1)}{2}$. That is

$$e(t) = e^{i\omega_o t} \frac{\sin(\frac{N\omega T}{2})}{\sin(\frac{\omega T}{2})} \quad (2.3)$$

The average laser power output is proportional to $e(t)e^*(t)$, and is given by

$$P(t) \propto \frac{\sin^2(\frac{N\omega T}{2})}{\sin^2(\frac{\omega T}{2})} \quad (2.4)$$

From equation (2.4) we can conclude: [1]. The power is emitted in a pulses train, with period $T = \frac{2\pi}{\omega} = \frac{2L}{c}$. [2]. The peak power is equal to N times the average power, where N is the number of modes locked together. [3]. The individual pulse width defined as $\tau = \frac{T}{N}$, and the number of oscillating modes can be approximated by the ratio of the transition lineshape width $\Delta\omega$ to the frequency spacing ω between the modes, $N = \frac{\Delta\omega}{\omega}$. and since $T = \frac{2\pi}{\omega}$, we have

$$\tau = \frac{2\pi}{\Delta\omega} = \frac{1}{\Delta\nu} \quad (2.5)$$

Thus the length of the mode-locked pulses is approximately the inverse of the gain linewidth.

The duration of the single pulse envelope of equation (2.4) is roughly the reciprocal of the bandwidth as we would expect from fourier theory. The time and the bandwidth product taking into account the Uncertainty Principle is given by

$$\Delta t \Delta \nu \geq K \quad (2.6)$$

where K is a constant of the order of unity, whose value depends upon the shape of the intensity profile. For a Gaussian pulse,[2.9]

$$K = \frac{2\ln(2)}{\pi} = 0.441 \quad (2.7)$$

The value of $\Delta t \Delta \nu$, called the "time-bandwidth product" is a most important parameter in the study of ultrashort pulses, and is used as a measure of the extent to which a perfect mode-locked situation has been achieved. If the time-bandwidth product of a pulse approaches the lowest limit set by equation (2.6), the pulse is said to be "bandwidth-limited" or "transform-limited", and the shortest pulse obtainable with a given spectral bandwidth is

$$\Delta t = K \cdot \frac{1}{\Delta \nu} \quad (2.8)$$

2.1.2 Methods of mode-locking

Mode-locking may be accomplished by several different methods. We classify these as : [1]. Active mode-locking, which includes loss and phase modulation, and synchronous pumping. [2]. Passive mode-locking, and [3]. Hybrid mode-locking.

2.1.2.1 Active mode-locking

Active loss and phase modulation involves an intra-cavity loss or phase modulator, namely acousto-optics or electro-optics driven by R. F. signals whose period is synchronized to an integer multiple of the cavity round trip time^{[2.9] [2.10]}.

In this case an external modulation signal is applied, and the period must be matched accurately to the transit time of the laser cavity. Loss modulation, phase modulation and gain modulation (synchronously pumping) can all be used. The simplest procedure involves applying the sinusoidal output of a R. F. signal generator

to an acousto-optic or electro-optic element in the cavity, or superimposing the R. F. signal on the driving current as in the case of semiconductor lasers. Actively mode-locking by sinusoidal modulation is reliable and the pulses are easily synchronised to other devices. On the other hand, because the R. F. modulation is relatively slow varying, the pulse durations generated are in the tens of ps regime.

The alternative technique that has been developed for generating a deeper profile of modulation involves modulating the gain of the laser by pumping with another mode-locked source of accurately matched cavity length, namely mode-locking by synchronous pumping. In synchronous gain modulation the leading edge of the pulse in the slave laser is suppressed by arranging the timing so that the rising gain in the active medium preferentially amplifies its peak. If this pulse has enough intensity it will deplete the gain and so the trailing edge will also be suppressed, leading to pulse shortening. This technique enables shorter pulse to be produced than other active loss and phase modulation. Many excellent theoretical and experimental investigations can be referred to in reference [2.9].

2.1.2.2 Passive mode-locking

In the case of passive mode-locking, mode-locked operation is achieved simply by inserting in the cavity a saturable absorber that is placed to the far end of the output coupler, near or simply in contact with the total reflecting mirror^[2.11]. In a multimode laser the optical signal in the absence of mode-locking consists of a sequence of random fluctuations. Since larger fluctuations cause greater saturation

than smaller ones they receive preferential transmission, so that in the optimal case, one fluctuation ultimately dominates all the others. The saturable absorber is usually an organic dye in the cuvette, flowing jet, or even in a plastic sheet. However colour centre crystals have also been used^[2.11].

The technique which is most commonly used in CW dye lasers was developed in 1972 by Ippen et al^[2.12]. The method has also been used in solid state laser systems such as Nd:glass lasers. Passive mode-locking is simple and inexpensive and is generally far more effective than active mode-locking for the generation of ultrashort pulses. In recent years, the availability of suitable saturable absorber and the use of harmonic generation has enabled almost all the UV to near IR spectrum regime be covered^[2.13].

The nonlinear processes of both saturable gain in the active (amplifying) medium and saturable absorption in the passive (modulating) medium are found to play an important role in passive mode locking in dye lasers. The leading edge of a pulse in the cavity is suppressed by the absorption in the saturable absorber before it saturates, and the trailing edge is suppressed by the gain saturation in the gain medium. The time evolution of the pulse compression has been analysed by New^[2.9]. He gives a description of how the leading edge of a pulse is suppressed by the absorption in the saturable absorber before it saturates, and the trailing edge of the pulse is suppressed by gain saturation in the lasing medium. It is necessary to adjust the gain in the active medium and the absorption in the passive medium such that the pulse is not able to saturate the absorber too easily yet it is still able to sufficiently deplete the

gain. According to New's model, this occurs when the laser is designed such that the absorption saturates before the gain, and the gain medium does not have time to fully recover between pulses.

As shown in the stability diagram in Fig. 2.2. The ratio of the effective absorption cross section to the effective gain cross section is given by the "S" parameter S [2.9]

$$S = K \frac{A_a \sigma_b}{A_b \sigma_a}$$

where A_a and A_b are the beam areas in the gain and absorption media respectively; σ_a , σ_b are the respective cross-sections of the stimulated emission and absorption; and K is a constant equal to unity. " g_0 " is the small signal gain in the system and " ξ " is given by the ratio of the cavity round trip time to the relaxation time of the active dye. Fig. 2.2 shows that for different values of " S ", there are boundaries of the regions where these conditions for pulse compression and hence stable pulse evolution are fulfilled. It also shows that for large values of " S ", the restriction on the pump power and the cavity length are less severe.

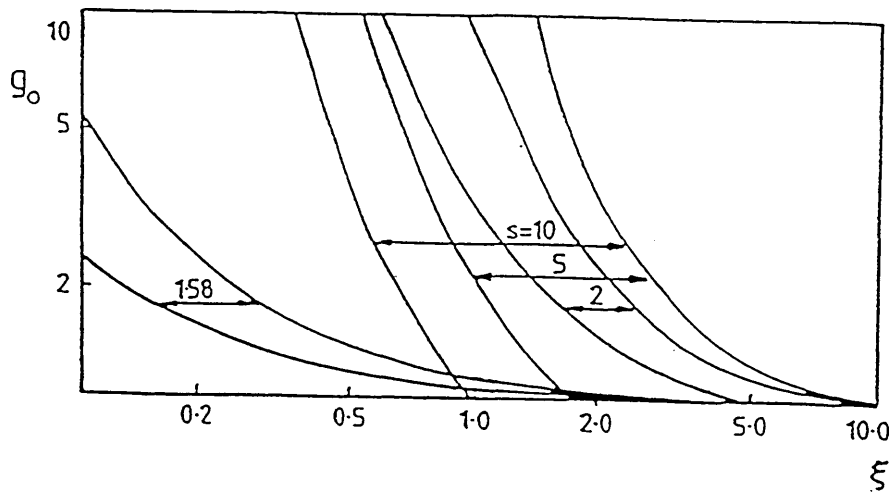


Figure 2.2 The stability diagram for passive mode-locking.
(Taken from reference [2.9])

Physically, the stability parameter measures the relative effectiveness of a photon in the laser cavity to saturate the absorber medium compared to saturating the gain medium. A stability parameter greater than 1 implies that loss occurs on both the leading and the trailing edges of the optical pulse with gain at the peak of the pulse. This condition is necessary for the formation of a short pulse in a passively mode-locked dye laser. To assure pulse forming stability, in designing a passively mode-locked dye laser cavity, the radii of curvature of the mirrors focusing into the absorber medium are chosen to be smaller than the radii of the mirrors focusing into the gain medium. This assures that the spot size for the laser mode is smaller in the absorber medium than in the gain medium.

Haus^[2.18] included a bandwidth limiting term in his theory of passive mode-locking and derived a pulse shape given by

$$I(t) = \text{sech}^2\left(\frac{t}{\tau_p}\right)$$

where τ_p is the pulse width inversely proportional to the bandwidth, the pulse energy and the square root of the small signal loss on the absorber.

The first experimental CW passively mode-locked dye laser^[2.12] used R 6G as the gain medium pumped by Argon ion laser 514 nm line, and DODCI as the saturable absorber. Both the active and passive media were situated in the folding section to increase the energy densities, and so increase the pumping efficiency of the system. Typical pulsewidths generated by passive mode-locking are in the picosecond and sub-picosecond regime.

2.1.2.3 Hybrid mode-locking

Another technique of mode-locking is a combination of both active and passive mode-locking, called hybrid mode-locking. In a typical configuration the dye laser is actively mode-locked (almost always via synchronous pumping) and still contains a saturable absorber in the cavity which has the effect of reducing the pulse wings and giving pulse duration up to an order of magnitude shorter than with active mode-locking alone.

Hybrid mode-locking was first demonstrated^[2.14] by Shank and Ippen in 1974 where they used both active and passive dyes (Rh 6G/DODCI) flowing in a single jet stream. Pulses with 0.5 ps to 1 ps duration were produced. Various active/passive dye combinations, cavities with separate flowing jets for active and passive dyes and intracavity compensating configurations have also been used for hybrid mode-locking and produced shorter pulses well down towards the femtosecond regime. This method can use a wide range of active/passive dyes, can easily be synchronised to other system and is capable of producing pulse durations of 29 fs^[2.15] almost as short as purely passive mode-locking. Yet its inferior pulse quality and stability when compared with purely passive systems are the disadvantages.

2.1.3 Ultrashort pulse generation

Mode-locking techniques enable pulses with durations in picosecond and sub-picosecond regimes to be generated routinely. But to generate ultrashort pulses with

durations less than a hundred femtoseconds or tens of femtoseconds more efforts are needed. This can be achieved by applying colliding pulse techniques in the passive saturable absorber and by properly compensating the intracavity group velocity dispersion.

2.1.3.1 Colliding pulse mode-locking (CPM)

The colliding pulse technique takes advantage of the fact that two counter-propagating pulses that collide in a saturable absorber jet create a transient grating^{[2.16][2.17]}. This reduces the energy needed to saturate the absorber by increasing the effective absorber cross section and so enables the generation of shorter pulses^[2.18]. In a CPM ring cavity configuration if the gain medium and the absorber medium are separated by a quarter of a cavity length, the pulses will arrive at the gain medium equally spaced in time, thus both pulses will see the same and maximum saturable gain, as shown in Figure 2.3.

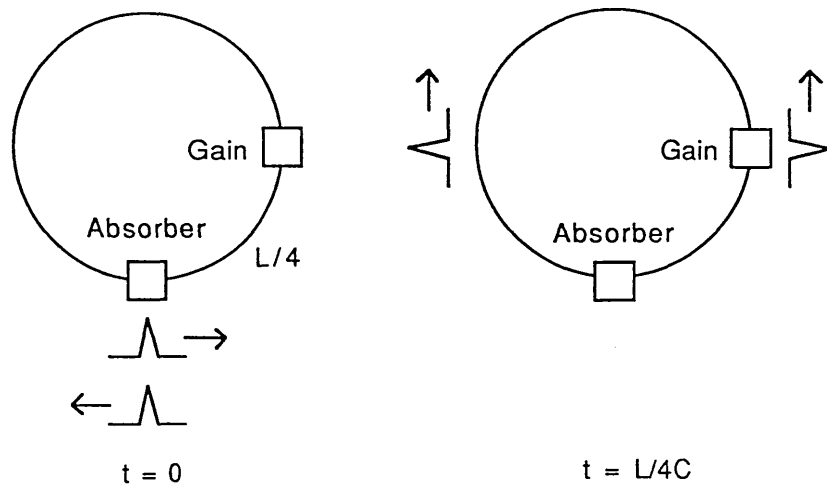


Fig. 2.3 $L/4$ cavity length spacing between the gain and absorber jet.

The first experiment using the CPM technique was with an Ar^+ ion laser pump Rh 6G/DODCI dyes combination that produced ultrashort pulses of 90 fs^[2.19]. There was no tuning or bandwidth limiting element and the laser ran at 627 nm. By changing the dielectric mirrors in the cavity, pulses down to 65 fs and 55 fs were produced. The basic CPM mechanism has been shown theoretically to produce pulse which are chirped according to the amount of detuning of the pulse frequency away from the centred frequency of the saturable absorber. The improvements in the above pulse durations were later shown to be due to the compensation of this chirp by balancing the total self phase modulation and the group velocity dispersion in the cavity. The shortest pulse duration directly generated from a CPM laser is 27 fs^[2.20]. The CPM interaction also improves mode-locking stability.

Compared with other techniques for producing ultrashort pulses, passive mode-locking has the advantages of producing the shortest pulses with good stability and low interpulse jitter.^[2.21] It is also cheaper than other techniques since no active components, driving devices or other mode-locked pump lasers are required.

2.1.3.2 Dispersion compensation

The often conflicting roles of group velocity dispersion (GVD) and self phase modulation (SPM) have become very important in the generation of ultrashort pulses. The shortest pulses of 27 fs from the laser cavity were generated by balancing intra-cavity self phase modulation, group velocity dispersion, saturable absorption and saturable gain^[2.19]. Furthermore, the shortest pulses ever of 6 fs were generated us-

ing more extracavity controlled group velocity dispersion and self phase modulation stages.^[2.6]

Group velocity dispersion mostly arises from optical elements of glass, quartz, dye jet and the reflection of the dielectric mirrors; it can be compensated by using diffraction grating pairs^[2.22], prism pairs^[2.23], and interferometers^[2.24].

Self phase modulation arises from the non-linear interaction of the intense light intensity with optical media. In the passive mode-locked dye lasers the main causes of SPM are negative SPM due to off resonant saturation of the saturable absorber which gives a down chirp, and positive SPM due to the optical Kerr effect in the solvents which gives an up chirp.^[2.25] The SPM arising from gain saturation is in general less significant due to the tighter focusing employed in the passive section of the laser. In the optical Kerr effect, the interaction of light produces a change Δn in the refractive index of the medium through which it travels given by

$$\Delta n = n_2 I \quad (2.9)$$

where n_2 is a medium dependent coefficient and I is the incident intensity. There is thus an intensity induced phase change given by

$$\Delta \Phi = \frac{2\pi}{\lambda} L \Delta n = \frac{2\pi}{\lambda} L n_2 I \quad (2.10)$$

where L is the length of material. If the intensity varies with time, then the phase change will be a function of time, so there will be a frequency chirp $d\omega$ given by

$$d\omega = \frac{d\phi(t)}{dt} = -\frac{2\pi}{\lambda} L n_2 \frac{dI(t)}{dt} \quad (2.11)$$

In an optical pulse the frequencies in the leading edge (where $\frac{dI}{dt}$ is positive) are thus lowered; and those in the trailing edge (where $\frac{dI}{dt}$ is negative) are thus raised giving an overall up chirp.

The unsaturated gain in the amplifier, and the unsaturated loss in the absorber give chirps that can be obtained by considering a Lorentzian lineshape centred at frequency ω_0 . The refractive index at ω is given by

$$n(\omega) = n_0 + \frac{\pi}{2}(\omega_0 - \omega)\frac{c}{\omega}g(\omega_0 - \omega)\alpha \quad (2.12)$$

where n_0 is the refractive index in which the transition medium is dissolved, c is the speed of light in vacuum, α is the gain coefficient at the transition peak, and $g(\omega_0 - \omega)$ is the Lorentzian lineshape given by

$$g(\omega_0 - \omega) = \frac{2}{\pi\Delta\omega_0}\left[1 + \left[\frac{2(\omega_0 - \omega)}{\omega}\right]^2\right] \quad (2.13)$$

where $\Delta\omega_0$ is the FWHM linewidth.

When an intense optical pulse saturates the absorber in a passively mode-locked laser, the absorption coefficient will have a time dependence which will thus cause a time dependent refractive index as in eq(12). This again gives a time dependent phase

$$\phi(t) = -\omega_0 n(t) \frac{L}{c} \quad (2.14)$$

which gives a frequency chirp as in eq(11), but with the opposite sign to the Kerr effect.

Practically these two non-linear effects are changed in the passively mode-locked lasers by moving the absorber away from the focus in the folded section because this changes the energy density and thus the intensity that the saturable absorber sees.

Similar SPM phenomena could have been considered in the active amplifying dye section as arising from the gain saturation. However, this effect can be ignored on account of the lower saturation of the amplifier due to the less tightly focusing and the greater spot size, and the absence of the grating formation in the gain medium.

The other main source of the intracavity dispersion is the optical components. The phase shift of an EM wave of the form $e^{i(\omega_0 t - KZ)}$, after traversing an effective length L of a transparent optical material is given by

$$\phi(\omega) = -\omega n(\omega) \frac{L}{c} \quad (2.15)$$

The second derivative of this expression is

$$\frac{d^2 \phi}{d\omega^2} = -\frac{L}{c} \left(2 \frac{dn}{d\omega} + \omega \frac{d^2 n}{d\omega^2} \right) \quad (2.16)$$

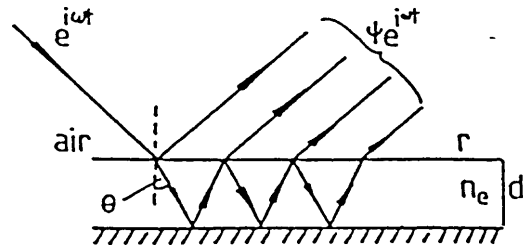
The dispersion due to these optically transparent materials, i.e. normal dispersion always gives a positive frequency chirp.

When coherent light is incident on a typical multi-layer dielectric laser mirror, the reflected wave is produced by the interference of the multiple reflections within the dielectric layers. Besides a frequency dependent amplitude modulation (i.e. reflectivity, $R(\omega)$), the dielectric layers also import a frequency dependent phase shift $\phi(\omega)$ to the reflected wave. Thus a dielectric mirror acts as a source of GVD whose dispersive power is given by $d^2 \phi / d\omega^2$. This function has been calculated by Silvestri et al^[2,24]. When a narrow-band dielectric mirror is used at its resonant wavelength, i.e. the centre of the high reflectivity curve, there is negligible group velocity dispersion. However, if the mirror is used off resonance, it does become dispersive, and the

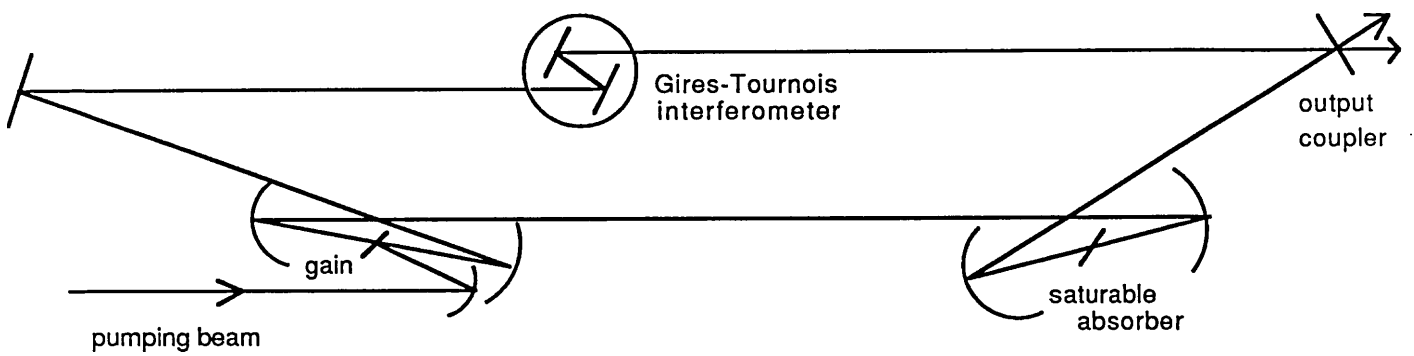
contribution to the cavity GVD could be equivalent in magnitude to that dispersion introduced by propagation through several millimeter of quartz^[2.24].

To generate ultrashort femtosecond pulses we need to compensate for all these dispersions. Generally, there are two intracavity dispersion compensating elements, namely, Gires-Tournois interferometer^[2.24] and a prism pair sequence as shown in Fig. 2.4^[2.23].

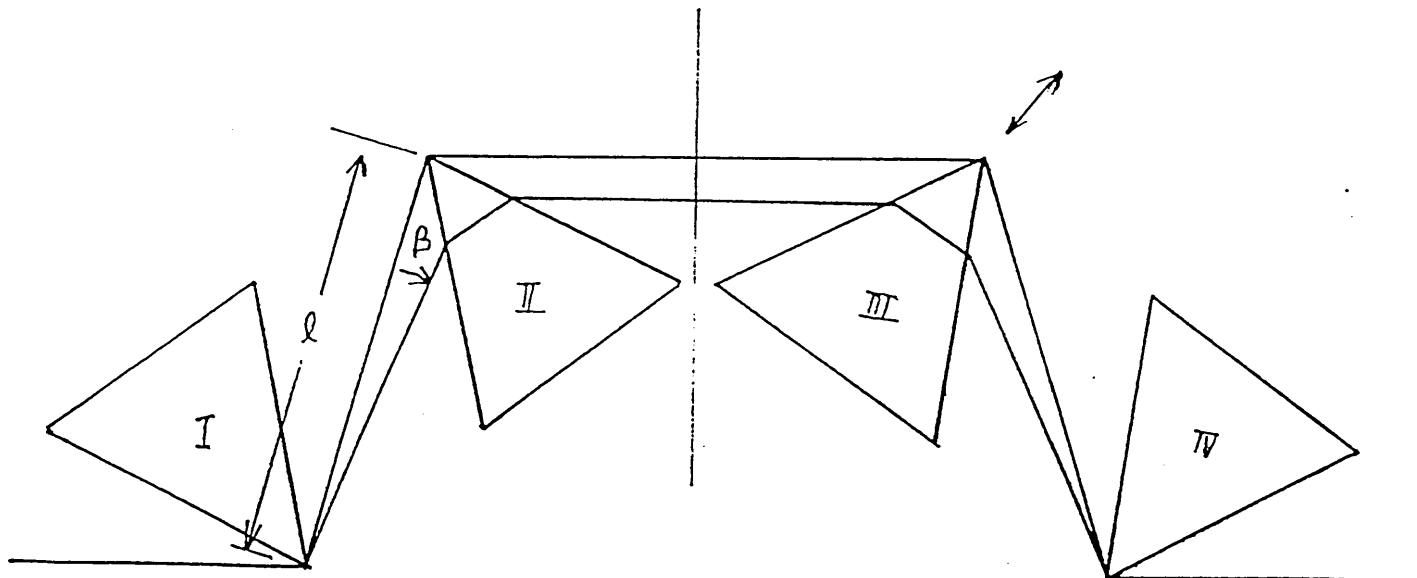
The prisms are used at minimum deviation and are cut so that the rays enter and leave at the Brewster's angle. The first prism introduces an angular dispersion of the incident beam into prism two so that the lower frequency components will see a longer optical path in the second prism than the higher frequency components. Hence the spectral components emerging from the second prism will have the same delay. The spectral components are all parallel to the incident beam but spatially distributed. The spatial spread may then be undone by passing the rays through another identical prisms pair, or in a linear cavity reflecting them back through the same prism pair again. The four prisms sequence is easy to employ into a ring cavity since there is no net displacement of the incident beam. The net GVD of the prism sequence is adjustable through positive values down to zero and negative values simply by properly separating the distance between P_1 and P_2 that gives the negative amount of GVD and by translating one of the prisms along an axis perpendicular to its base since thus controls the amount of positive GVD.



(a) Schematic of Gires-Tournois interferometer



(b). Colliding pulse mode-locked ring dye laser using Gires-Tournois interferometer as dispersion compensating element.



(c) Intracavity GVD compensating prism pairs.

Fig. 2.4 Gires-Tournois interferometer and prism pairs configuration.

The Gires-Tournois interferometer which is essentially an etalon that has 100 per cent reflectivity on one surface gives adjustable linear or non-linear GVD. It has the advantages over prism sequences of being potentially more dispersive, more compact, applicable to all polarisations and over a wide range of frequencies^[2.27].

It has generally been found that lasers with a small amount of excess negative GVD produce the shortest and most stable pulses^{[2.19] [2.28][2.29]}, where as those with excess positive GVD tend to give broader and unstable pulses. The shorter pulses do of course require larger bandwidths, and to generate the shortest pulses it will be necessary to compensate for the higher order terms of dispersion. By properly balancing the SPM, GVD, saturable absorption and saturable gain^[2.30], direct generation of optical pulses as short as 27 fs from a laser has been achieved^[2.19].

If a pulse is not bandwidth limited, i.e. has chirps, then adjusting the GVD by an appropriate amount will "unchirp" it and thus narrow the pulse down to its bandwidth limit. The prisms sequence and the Gires-Tournois interferometer mentioned above for intracavity operation can also be used extracavity. However, this is less effective because the pulse traverses only once instead of many times as inside a laser cavity.

Another important device that can give negative GVD is a pair of diffraction gratings^[2.22]. The spatial separation of spectral components is introduced and used to produce the temporal shift, in the same way that the dispersion is introduced in the prism sequence. The emerging beam is reflected back through the grating so that it emerges parallel to the incident beam, with the dispersion dependent on the

grating separation, the angle of incidence, the frequencies of the input beam and the grating constant. The negative dispersion for a typical set up with 1m grating separation is usually several orders of magnitude larger than that obtained with prism pairs or dielectric surfaces. The main applications for the grating pair has been in the fibre-optic pulse compressor^[2.31] where a pulse is first frequency broadened and linearly chirped by passing through an optical fibre then compressed by the diffraction gratings dispersive delay line. However, because of its relative high loss the grating pair can not be used inside CW dye lasers.

A combination of a grating pair and a prism sequence has been used to compensate for both quadratic and cubic phase distortion of the pulse, and it is this combination that has produced the shortest pulse to-date, 6fs.

2.2 Ultrashort pulse measurement

The invention of the passively mode-locked Nd:glass laser in 1965^[2.4] resulted in a pressing need for new techniques to measure the duration of shorter optical pulses.

2.2.1 Direct measurement of ultrashort pulses

There exist several linear techniques for directly measuring temporal intensity profiles. One of the simplest involves the use of a photodiode to convert an optical signal into an electrical signal, and an oscilloscope to display it. Even if the signal is repetitive, as in CW mode-locked dye lasers, and pulse sampling techniques are used,

the resolution of this combination will still be limited by the rise time of the sampling head. The fastest photodiode reported has a deconvolved resolution of 16 ps. Ultrafast semiconductor switch/oscilloscope combinations suffer for the same reason with convolved resolutions of the order of 40 ps being readily obtained^[2.33]. Non-conventional sampling techniques that use arrays of semiconductor switches have been developed with overall time resolutions of 10 ps^[2.34]. However, for direct measurements of pulses of 1 ps duration an electron-optical streak camera must be used.

The streak camera is the only direct measurement device capable of picosecond and sub-picosecond resolution^{[2.35][2.36]}. The principle of the streak camera was first proposed by E. K. Zavoiski et al in 1956^[2.8], but it was only after 1969 that it became a potential picosecond measurement system^[2.37]. The optical signal is imaged from a slit onto a photocathode that emits electrons. The number of electrons emitted is proportional to the light intensity hitting the photocathode. The electrons are accelerated through electrodes and deflection plates and focused onto a phosphor. By applying a fast synchronised ramp signal to the deflector the signal is streaked across the phosphor screen. The time information of the optical signal is thus converted into a temporal distribution of the electron signal which in turn is converted into a spatial intensity distribution on the screen. This intensity pattern may then be analysed by the densitometer from the films or by the CCD device and computer immediately to display the temporal profile of the incident signal. For the Photochron II the writing speed is $2 \times 10^8 \text{ ms}^{-1}$, and the spatial resolution is 10^4 lines/m, giving a technical time resolution of 0.5 ps. Advances in photocathode and deflection plate design has improved the time resolution of streak cameras to 0.3 ps^[2.36].

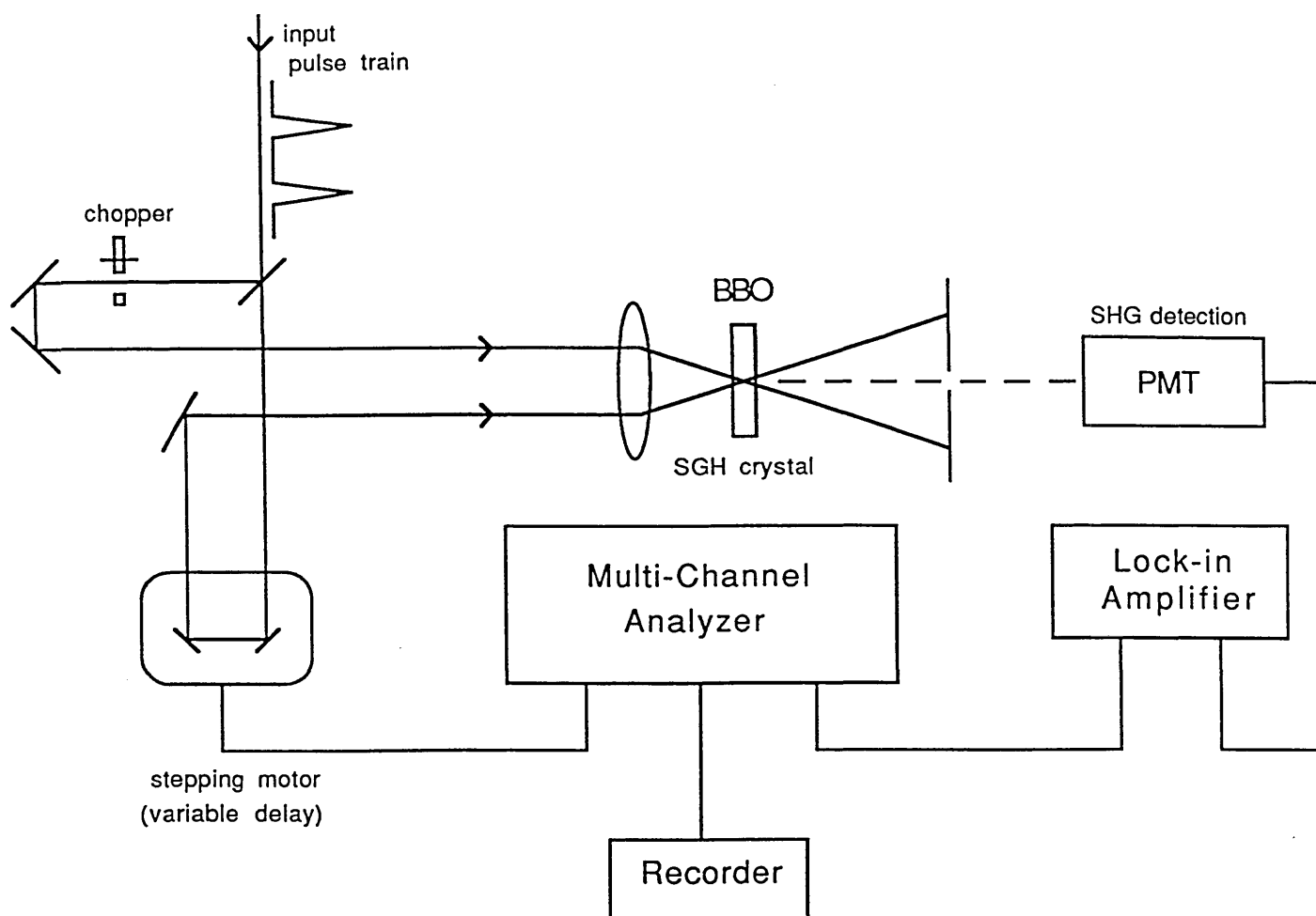
Direct measurement by the combined use of photodetectors and oscilloscopes was no longer adequate to resolve the pulses produced temporally. Even up to recently, the best direct pulse measurement by this photodiode-oscilloscope combination using pulse sampling techniques could only measure sub-hundred picosecond. Nevertheless, it still provides a very good monitor on the mode-locked pulse train. Recently, commercial available streak camera system can measure pulses of 1 ps reliably, and x-ray streak camera that detect UV pulses could also reach this range.

2.2.2 Autocorrelation measurement of ultrashort pulses

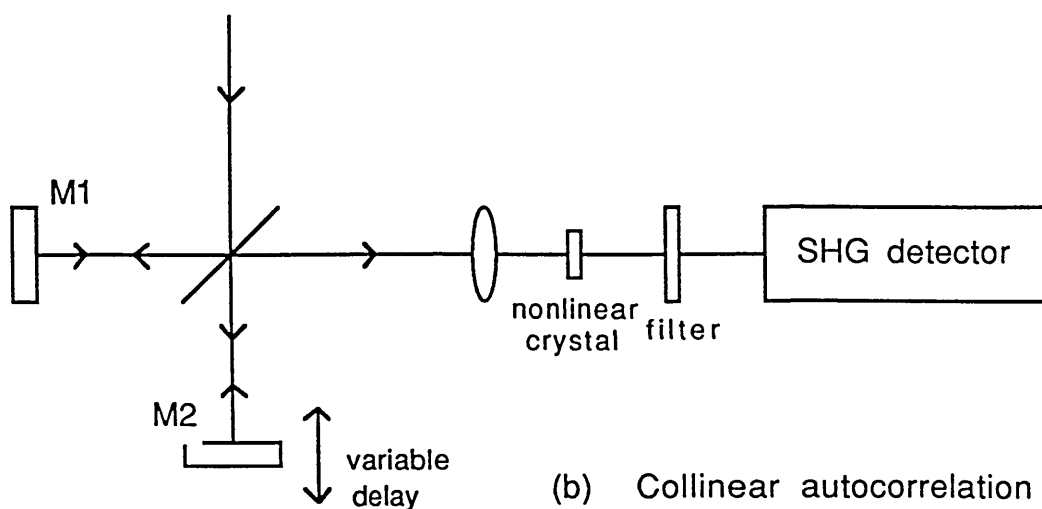
The first technique based on the nonlinear process of second harmonic generation (SHG) for the measuring of ultrashort pulsewidths with picosecond resolution was proposed by Maier et al. in 1966^[2,38]. Today this method still remains the most widely used method for characterising ultrashort pulses, and has been used to measure pulses as short as 6 fs corresponding to less than 4 optical cycles. An alternative nonlinear method for measuring ultrashort pulse durations is that of two photon fluorescence (TPF) which was developed^[2,39] soon after the SHG technique, and largely superseded the SHG technique for low repetition rate lasers. Both of these techniques yield the autocorrelation function of the pulse intensity profile.

Optical SHG occurs when two photons combine in a nonlinear medium to produce a single photon of twice the original frequency. This process is enhanced only if the fundamental and second harmonic radiation are phase matched in a non-centrosymmetric nonlinear medium. A typical configuration for measuring pulse du-

ration using SHG autocorrelation is shown in Fig. 2. 5.



(a) Background-free autocorrelation.



(b) Collinear autocorrelation

Fig. 2.5 Typical configuration of SHG autocorrelation set up.

The pulses to be measured are divided into two beams of roughly equal intensity which are recombined in a SHG crystal after a relative delay has been introduced between them. By monitoring the average power of the second harmonic of the recombined beam produced as the relative delay between the two pulse trains are varied, the autocorrelation function of the incident pulse can be obtained. When the fundamental field is

$$e_1(t) = \text{Re}[E_1(t)e^{i\omega t}] \quad (2.17)$$

the second harmonic field is

$$e_2(t) = \text{Re}[E_2(t)e^{i2\omega t}]$$

and

$$e_2(t) \propto \text{Re}[E_1^2(t)e^{i2\omega t}] \quad (2.18)$$

The input pulses pass through a 50/50 beam splitter and one of the beams is delayed by τ relative to the other, so that

$$\begin{aligned} e_{total}(t) &= \text{Re}\left[\frac{1}{2}E_1(t)e^{i\omega t} + \frac{1}{2}E_1(t-\tau)e^{i\omega(t-\tau)}\right] \\ e_{total}(t) &= \text{Re}\left[\frac{1}{2}E_1(t) + \frac{1}{2}E_1(t-\tau)e^{-i\omega\tau}\right]e^{i\omega t} \end{aligned} \quad (2.19)$$

Setting

$$E(t) = E_1(t) + E_1(t-\tau)e^{-i\omega\tau} \quad (2.20)$$

it follows that

$$e_{total}(t) = \frac{1}{2}\text{Re}[E(t)e^{i\omega t}] \quad (2.21)$$

Since

$$e_2(t) \propto [E(t)]^2 = [E_1(t) + E_1(t-\tau)e^{-i\omega\tau}]^2 \quad (2.21)$$

then

$$e_2(t) = E_1^2(t) + E_1^2(t - \tau)e^{-2i\omega\tau} + 2E_1(t)E_1(t - \tau)e^{-i\omega\tau} \quad (2.22)$$

and the photo-current generated in the photomultiplier (PMT) with a narrow band pass filter to cut the fundamental radiation and pass only the second harmonic radiation is

$$i(t) \propto E_2(t)E_2^*(t) \quad (2.23)$$

and

$$\begin{aligned} i(t) \propto & [E_1^2(t) + E_1^2(t - \tau)e^{-2i\omega\tau} + 2E_1(t)E_1(t - \tau)e^{-i\omega\tau}] \\ & \times [E_1^2(t) + E_1^2(t - \tau)e^{2i\omega\tau} + 2E_1(t)E_1(t - \tau)e^{i\omega\tau}] \end{aligned} \quad (2.24)$$

so

$$i(t) \propto [E_1(t)E_1^*(t)]^2 + [E_1(t - \tau)E_1^*(t - \tau)]^2 + 4E_1(t)E_1^*(t)E_1(t - \tau)E_1^*(t - \tau) + S(\tau) \quad (2.25)$$

The term $S(\tau)$ contains harmonic functions in $\omega\tau$ and $2\omega\tau$, which tend to average to zero by integration in the PMT as the delay is scanned, unless an extremely slow trace is taken, resulting in an interferometric autocorrelation which resolves the interference fringes between the beams in a collinear geometry.

The PMT cannot resolve oscillations on a timescale of t . It averages these out to a DC level. Terms in τ can be collected in terms of the intensity I , given by EE^* , and

$$i(\tau) \propto \langle I^2(t) \rangle + \langle I^2(t - \tau) \rangle + 4\langle I(t) \rangle \langle I(t - \tau) \rangle \quad (2.26)$$

after normalisation, where

$$G^2(\tau) = \frac{\langle I(t)I(t - \tau) \rangle}{\langle I^2(t) \rangle} \quad (2.27)$$

then

$$i(\tau) \propto 1 + 2G^2(\tau) \quad (2.28)$$

$G^2(\tau)$ represents the well known second order correlation function, and is non-zero when pulse overlaps with itself. A photocurrent is enhanced when the delay is adjusted such that the two pulses overlap temporally in the SHG crystal. The pulse duration is related to the magnitude of the delay range $\Delta\tau$ over which an enhancement in the SHG photocurrent occurs.

For $\tau = 0$,

$$i(\tau) \propto 1 + 2G^2(0) = 1 + 2 \times 1 = 3 \quad (2.29)$$

and for $\tau \gg \tau_0$,

$$i(\tau) \propto 1 + 2G^2(\infty) = 1 + 2 \times 0 = 1 \quad (2.30)$$

Equation (2.28) is true for the case of the collinear autocorrelation technique as shown in Fig. 2.5, where type I phase matching is employed, as both fundamental beams are of the same polarisation. This type of SHG is described as SHG of the first kind. In some experimental configurations, where the two beams have orthogonal polarisation,^{[2.39][2.40]} or they are parallel but do not propagate collinearly in the SHG crystal,^[2.38] the second harmonic signal is only obtained when the pulses are overlapping in the crystal. This is known as SHG of the second kind. The resulting intensity autocorrelation function is given by

$$i(\tau) \propto G^2(\tau) \quad (2.31)$$

The main advantage of using SHG autocorrelation of the first kind to measure

the ultrashort pulses is that the peak to background contrast ratio gives important information on the degree of mode-locking. This is illustrated in Fig. 2.6. For the perfect mode-locked pulses the contrast ratio is 3 : 1; for a free running laser with randomly phased modes the autocorrelation function has a contrast ratio of 3 : 2. Since for large τ where $I(t + \tau)$ and $I(t)$ are independent variables,

$$G^2(\tau \rightarrow \infty) = \frac{\langle I(t) \rangle^2}{\langle I^2(t) \rangle} = \frac{1}{2} \quad (2.32)$$

and $G^2(0)$ is by definition unity, we see that even a random signal produces a peak in its autocorrelation with $\frac{G^2(0)}{G^2(\infty)} = 2$. The width of this peak is a measure of the temporal coherence of the signal, and is just related to the inverse of the spectral bandwidth of the sources. For the partially or incomplete mode-locked pulses a contrast ratio of 3 : 2 : 1 is obtained. Since $I(t)$ could be described as $I(t) = I_1(t) \cdot I_2(t)$, where $I_1(t)$ is the intensity produced by a continuous source of thermal noise, and $I_2(t)$ is a more slowly varying envelope function of partially mode-locked signal, then

$$G^2(t) = G_1^2(\tau) \cdot G_2^2(\tau) \quad (2.33)$$

where $G_1^2(\tau)$ and $G_2^2(\tau)$ are the autocorrelation functions of $I_1(t)$ and $I_2(t)$ respectively. From an initial value ($\tau = 0$) of 1, $G^2(t)$ falls to a value of $\frac{1}{2}G_2^2(\tau)$ for τ longer than the coherence time of the random variable $I_1(t)$ and then to zero as $G_2^2(\tau) \propto \langle I_2(t)I_2(t + \tau) \rangle \rightarrow 0$. SHG of the second kind is also known as the "background free" autocorrelation technique, because it produces the best contrast ratio of 2 : 1, with no background, and is considered to give a better indication of the pulse wings.

After measuring the FWHM of the autocorrelation function $\Delta\tau$, the pulse duration Δt may be deduced once a particular pulse shape has been assumed. The second order autocorrelation function that is necessarily symmetric, does not uniquely define a temporal profile. Pulse shapes are inferred in practice by fitting the experimental autocorrelation traces to the theoretical profiles calculated for a variety of pulse shapes. Table 2.1 gives the $\frac{\Delta\tau}{\Delta t}$ ratio for various pulse profiles and the appropriate time-bandwidth products K for the transform limited pulses.

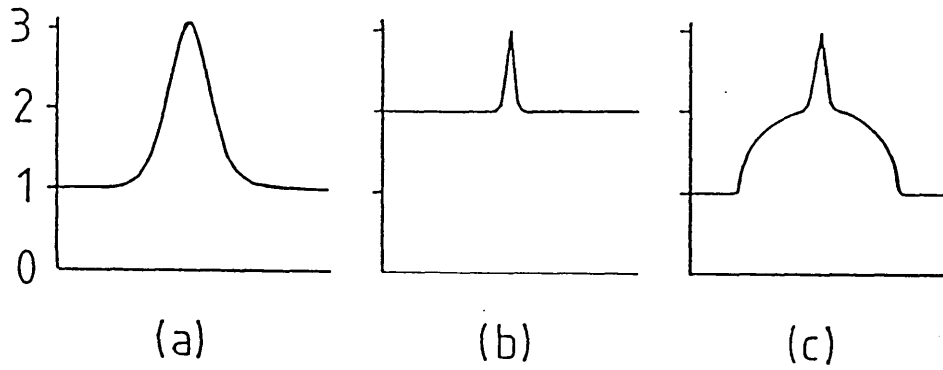


Fig 2.6 Autocorrelation traces resulting from: (a) a mode-locked laser, (b) a free running laser, (c) a partially mode-locked laser.

Table 2.1 Correlation widths and spectral bandwidths for four different transform-limited pulse shapes Δt and $\Delta\tau$ are full widths at half maximum of $I(t)$ and $G^2(\tau)$, respectively. $\Delta\nu$ is the FWHM of the measurable frequency spectrum.

$I(t)$	$\Delta\tau/\Delta t$	$K = \Delta t\Delta\nu$
$1(0 \leq t \leq \Delta t)$	1	0.886
$\exp[-\frac{(4 \ln 2)t^2}{\Delta t^2}]$	$\sqrt{2}$	0.441
$\text{sech}^2[\frac{1.76t}{\Delta t}]$	1.55	0.315
$\exp[-\frac{(\ln 2)t}{\Delta t}](t > 0)$	2	0.110

A powerful refinement of the SHG autocorrelation technique is that of interferometric autocorrelation^[2.41]. The experimental set up is similar to that described previously, but the relative delay between the two beams is adjusted with sufficient accuracy to resolve the interference fringes. As shown in equation (2.25), if the trace is taken slowly enough, $S(\tau)$ does not average to zero, and the fringes occur within the coherence time of the overlapping pulses. This type of autocorrelation gives an envelope of the constructive interference fringes with the peak to background ratio of 8 : 1 and is much more sensitive to the pulse shape and the frequency chirp of the pulse.

The technique of SHG autocorrelation is widely used because of its high temporal resolution - down to 6 fs. Yet temporal resolution will be limited by the finite phase matching bandwidth of the nonlinear crystal used, which results from the group velocity mismatch between the fundamental and the second harmonic radiation. Pulses with a spectral width in excess of this bandwidth no longer generate a second harmonic intensity signal proportional to the square of the fundamental intensity as derived in equation (2.28). In general, this could be avoided by using very thin SHG crystals as the phase matching bandwidth is inversely proportional to the interaction length^[2.42], or alternatively, by using non-collinear phase matching geometries such that two beams interact only over a short distance within the crystal. For a 100 μm thick KDP crystal, at 600 nm, the phase matching bandwidth is adequate to permit a resolution of 30 fs in a collinear geometry. A BBO crystal of 100 μm thick could also permit a temporal resolution of less than 50 fs in the shorter wavelength regime^[2.43].

In our system, in order to obtain the shortest seed pulse, a "real-time" autocorrelator is most useful. Fig. 2.7 is a schematic of the autocorrelation apparatus used. Mirror M_2 is attached to a shaker, (Bruel and Kjaer Ltd., Mini-shaker type 4810) which, when driven by a sinusoidal signal of approximately 6W output average power moves the mirror back and forth in a linear motion over as much as 5 mm. The motion of this mirror scans the pulses from one beam at the SHG crystal through their counterparts in the other beam, resulting in a repetitive second harmonic signal which may be displayed on the oscilloscope. This could be readily achieved by adding the signal from the PMT to a square wave which was synchronised with the sinusoidal signal driving the shaker. The PMT signal is directly proportional to the SHG signal because the fundamental signal is blocked by the narrow band-pass filter. The oscilloscope was triggered by the rising edge of the square wave and by adjusting the proper time base the complete autocorrelated pulse could be displayed in "real-time". This is most convenient and necessary when adjusting all the dispersion compensation to generate the shortest pulse and operate the laser within the stable regime. The oscilloscope display of the autocorrelated pulse could be calibrated by inserting a glass slide of known thickness into one arm of the interferometer and measuring the displacement of the peak of the autocorrelation trace.

It is also necessary to be able to measure the ultrashort pulse duration of UV laser sources at low repetition rates. In 1981 D. M. Rayner et al^[2.44] demonstrated that, by using multiphoton ionisation as the nonlinear process in the autocorrelation technique, ultraviolet laser pulses of 400 ps duration could be measured at 337 nm. Later on, in 1986 O. L. Bourne and A. J. Alcock^[2.45] showed that a true single shot

autocorrelator based on multiphoton ionisation could measure pulse durations with a lower limit of 25 ps. The major break through was when M. H. R. Hutchinson et al^[2.46] employed fluorescence from xenon excimers excited by two photon absorption of atom pairs, to measure pulses of 350 fs duration at 248 nm. This is a relatively simple and reliable method for measuring the durations of individual subpicosecond UV pulses. The technique is also applicable to ArF laser sources with a resolution of less than 100 fs. Single shot measurement of subpicosecond KrF pulse width by three-photon fluorescence of XeF visible transition has also been demonstrated^[2.47], and used to measure pulse widths of 220 fs. A single shot autocorrelator for KrF subpicosecond pulses based on two photon fluorescence of cadmium vapor at 508 nm has also been shown^[2.48].

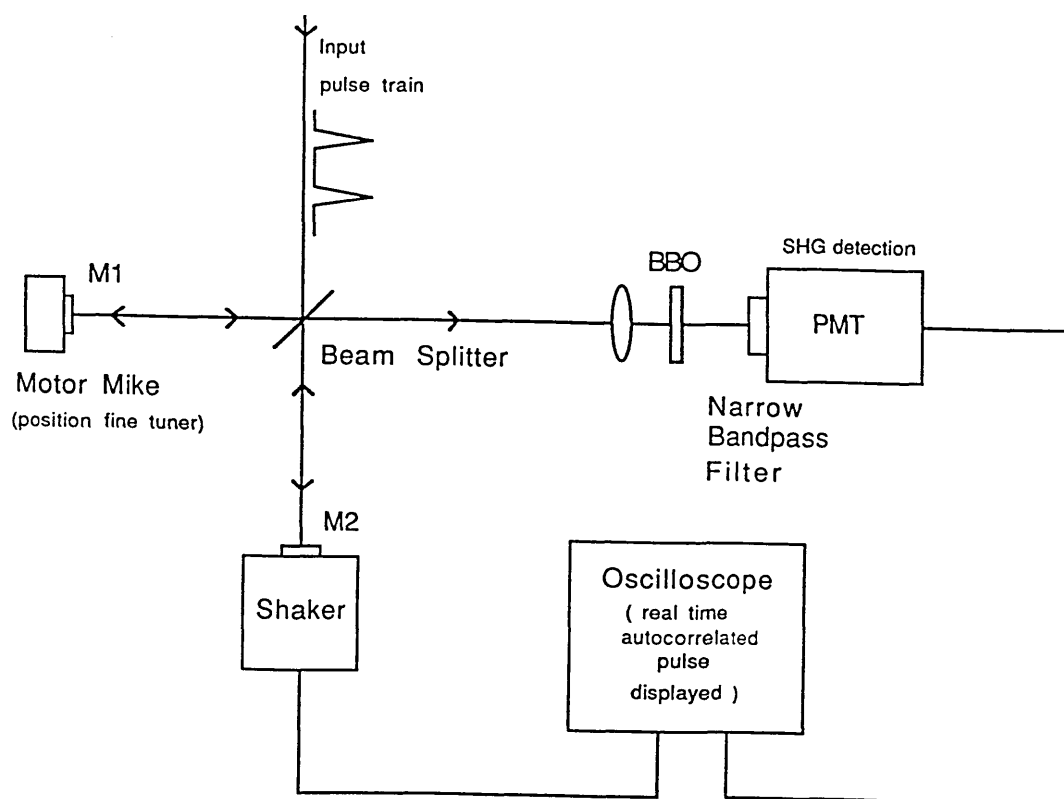


Fig. 2.7 Real time collinear autocorrelator.

Another approach to the measurement of intense UV subpicosecond pulses using degenerate four wave mixing, a back ground free measurement of the third order intensity autocorrelation function of XeCl excimer amplified pulses of 180 fs duration has been demonstrated^[2.49]. By using a tilted pulse front geometry and three photon fluorescence of XeF as the nonlinear detector, with CCD and oscilloscope as the readout system single shot UV pulses of 380 fs pulses have also been displayed with the best resolution to date of a few femtoseconds^[2.50].

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Chapter 3

CPM Ultrashort Pulse Blue-Green Dye Laser

Our system initially requires ultrashort pulse generation from a seed oscillator. Dye lasers have broad bandwidths which can therefore support ultrashort pulse generation. Direct generation of 27 fs pulses with a R 6G/DODCI dye laser in 627 nm have been demonstrated^[3.1]. From the previous investigations a mode-locked dye laser in a colliding pulse ring configuration with proper intracavity dispersion compensation elements is the best candidate. It is necessary to select the active(gain)/passive(absorber) dye combination that will lase at 497.2 nm which is twice the KrF excimer laser operating wavelength of 248.6 nm. Coumarin 102 active laser dye which can lase in the blue-green regime, from 480 nm to 510 nm; with the saturable absorber DDCI which has peak absorption at 496 nm is considered to be the best combination to achieve 497.2 nm. Using this combination together with an intracavity prism quadruplet for dispersion control^[3.2] and having optimized the operating conditions, we have achieved better than what has previously been reported^[3.3] by generating an 88 fs duration ultrashort pulse train at 497.2 nm with output power over 10 mW and tunable from 492 nm to 503 nm.

3.1 CPM ring dye laser

The ultrashort seed pulse is generated from a colliding pulse mode-locked ring dye laser. In this section the set up is described in full detail, with particular emphasis on the alignment techniques of the CPM system.

3.1.1 Configuration of the CPM ring dye laser

Fig. 3.1 shows the schematic set up of the CPM laser. An Ar^+ laser (Coherent, innova 100 series) with UV all lines output from 330 nm to 360 nm, is used to deliver up to 6 W as the pumping source. The CPM ring laser basically consists of an active dye jet with optical folding (M_1, M_2 and M_3), passive dye jet with folding (M_4 and M_5), prisms quadruplet, deflecting mirror M_7 , output coupler M_6 and the wavelength tuning slit A.

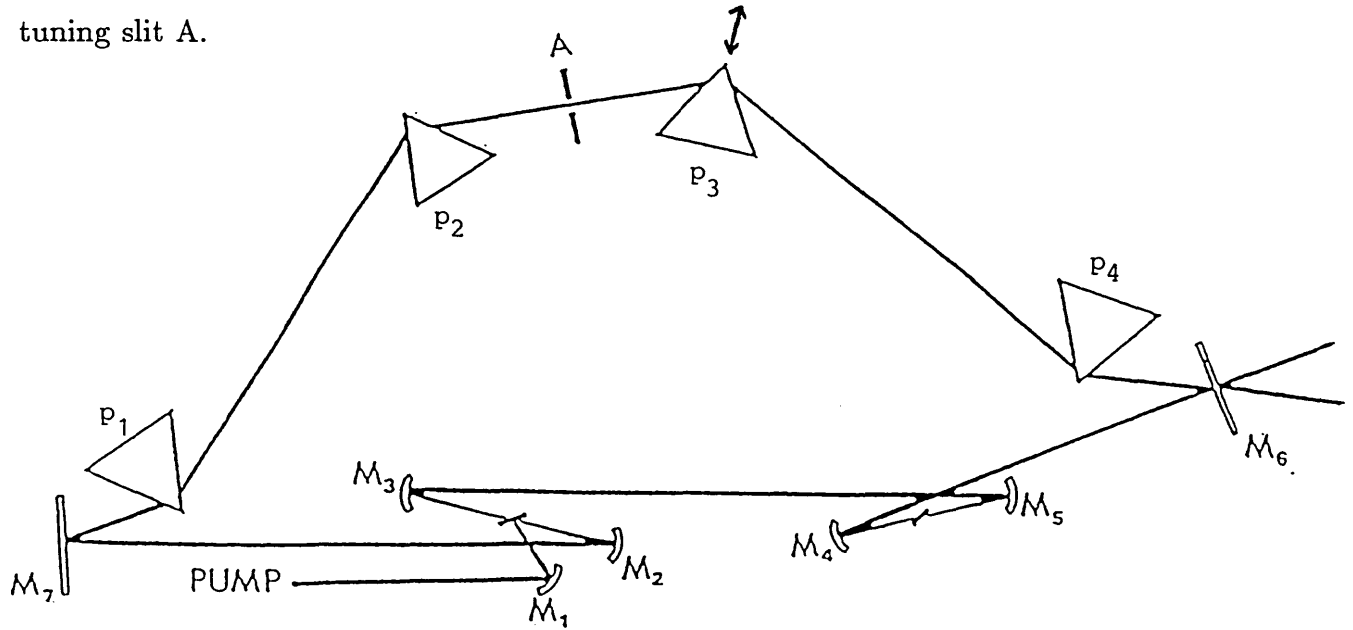


Fig. 3.1 C102/DOCI CPM ring dye laser configuration.

The active dye jet consists of a standard Coherent thick nozzle of 400 μm , resonant mirrors, and a pumping beam focusing mirror. The Coumarin 102 dye of $4 \times 10^{-3} M$ (1.5 g of Coumarin 102 powder dissolved in 300 cc of benzyl alcohol, and diluted in 1.2 L of ethylene glycol) concentration in the reservoir bottle sits in a temperature stabilized bath. The dye circulates through the nozzle to generate a sheet of stream. The circulation system was pumped by a mini-pump through the housing of the microfibre filter and a regulator. The resonant mirrors are a pair of curved mirrors of 50 mm radius of curvature, with a single stack high reflection dielectric coating centred on 490 nm. The Ar^+ pumping beam is focused to the dye jet using a separate focusing mirror of 20 mm focal length with high reflection coating on 330 nm to 370 nm at 45° incidence. This gives much greater flexibility in the alignment.

The passive dye jet has a separate but similar circulation system, the nozzle is 100 μm thick and the resonant mirrors are of 25 mm radius of curvature. The saturable absorber is DOCl and has a concentration of about $2 \times 10^{-5} M$ depending on the operating conditions.

The colliding pulse mechanism is achieved by the ring cavity configuration in the saturable jet. The ring cavity is formed by a single stack plano high reflecting mirror centred at 490 nm, which deflects the beam from the gain jet fold at a small angle through the prisms quadruplet. The beam is finally deflected back to the saturable absorber jet fold and partially transmitted (1 percent output transmittance) by the output coupler M_6 , and this completes the ring configuration.

A horizontal translation slit A in the centre of the prism sequence is used as

a wavelength selection element. It is simple, inexpensive and is not an intracavity bandwidth limiting component.

3.1.2 Alignment of CPM laser

To align the CPM ring cavity it is usually to start with a typical 3 mirrors cavity, and then a 4 mirror cavity for the conventional CW lasing. The saturable absorber fold section for the mode-locking modulation is added in a 6 mirror configuration. The total reflector and the output coupler, are tilted and the prisms sequence to control the intracavity dispersion compensation is inserted. This forms a ring cavity with colliding pulse action at the saturable absorber jet.

The 3 mirror cavity is shown in Fig. 3.2. The C102 gain jet fold composes of two HR mirrors and an output coupler with active jet in the confocal position of the HR mirrors. The Ar^+ all lines UV pumping beam is directed by a total reflector and then focused to the active jet via mirror M_1 . The fluorescence from the C102 gain jet is collected by the resonant mirror pair M_2 and M_3 . M_2 collects the fluorescence at normal incidence and totally reflects all the fluorescence back to M_3 . M_3 deflects the fluorescence to the output coupler M_6 with partial transmission/reflection thus forming the complete cavity.

For the further extension to the ring cavity, it is necessary to align the 3 mirror cavity to the 4 mirror with the extra total reflector M_7 in use. The 4 mirror cavity is show in Fig. 3.3. It is beneficial to align the fundamental gain section in this "Z"

fold because by properly aligning the folding, the astigmatism resulting from the off axis mirrors M_2 and M_3 and from the Brewster angled jet could be compensated^[3,4]. This reduces the minimum lasing threshold.

To include the passive saturable absorber section mirror M_5 is inserted between M_3 and the output coupler M_6 , and is tilted to reflect the fluorescence at a small angle. M_4 is aligned so that it collects the fluorescence from M_5 and reflects the fluorescence past the edge of M_5 onto the output coupler M_6 . Thus a linear 6 mirror cavity has been set up that includes a gain jet folding and a saturable absorber jet folding as shown in Fig. 3.4. The "crossed" arrangement of the saturable absorber jet fold can optimally compensate the astigmatism and coma aberration in the folding mirrors^[3,5].

The final step in the set up of the CPM ring laser is to form a ring cavity so that the counter-propagating beams can collide with each other. By the employing a four prism sequence a ring cavity can easily be aligned without any extra reflecting mirrors and the prism sequence serves well as an intracavity dispersion compensating element. We first tilt M_7 by a small angle and move in prism P_1 so that the fluorescence passes the apex of P_1 at the Brewster's angle and with minimum deviation. The beam refracted after P_1 is dispersed. Prism P_2 is moved in to the beam in the opposite direction to P_1 , so that the dispersed beam restores its parallel wavefront and is spectrally separated. The distance between P_1P_2 is an important parameter that determines the positive or negative value of the GVD. The beam after P_2 is restored by another pair of prisms P_3 and P_4 in a symmetric

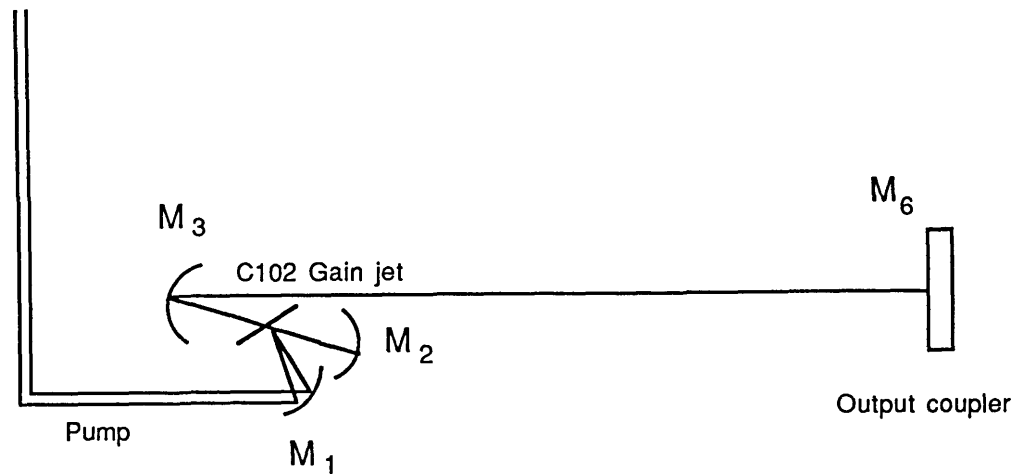


Fig. 3.2 Three mirror cavity.

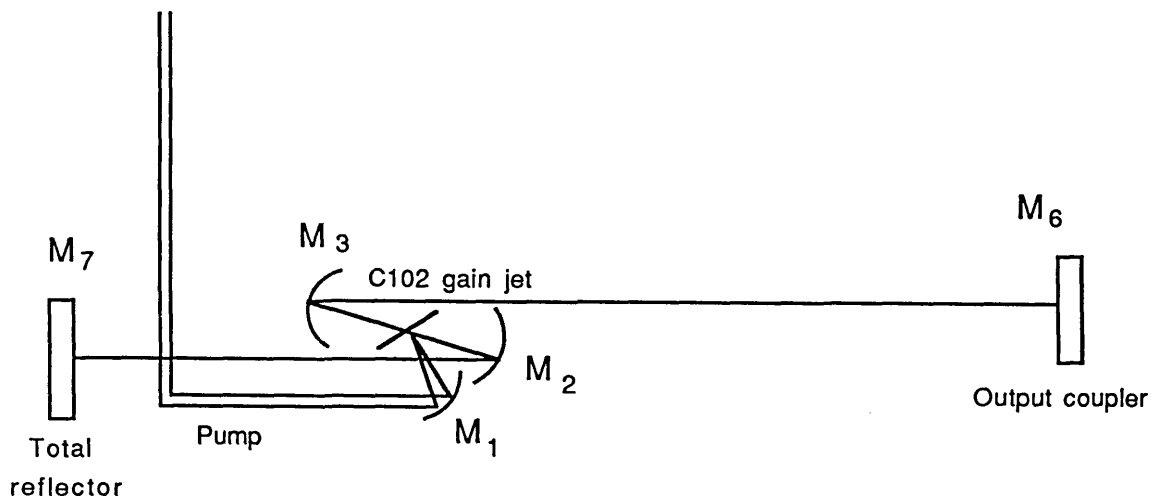


Fig. 3.3 Four mirror cavity.

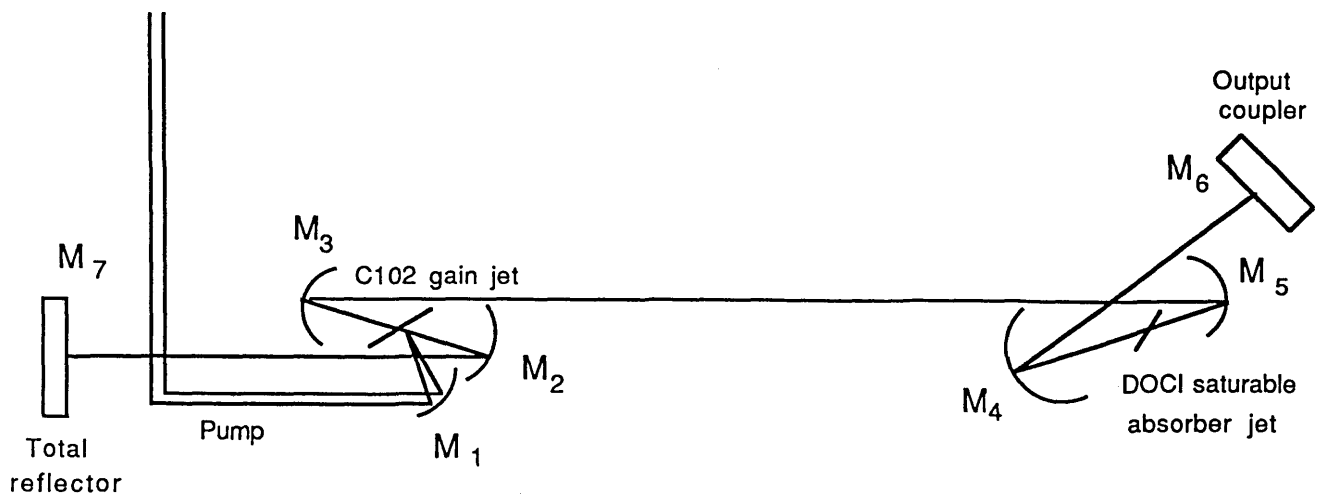


Fig. 3.4 Six mirror cavity.

configuration and this constitutes a complete ring cavity. One of the prisms, typically P_2 or P_3 because of its larger separation from other components and therefore reduced chance of interfering with the beam path, is mounted on top of a translation stage so that it can move perpendicularly to the prism base line to introduce more or less glass length for the intracavity dispersion compensation without changing the alignment.

A horizontal translating slit, A, with variable width could be inserted between prisms P_2 and P_3 as a wavelength tuning element.

3.2 Optimisations of the CPM ring dye laser

The alignment of a passively mode-locked ring dye laser has just been described. However, for the generation of ultrashort pulses several optimisations need to be done to various components, including the C102 active gain jet section, the DOCI passive saturable absorber jet section, the quarter length separation of the gain jet and the saturable absorber jet in the ring cavity, dispersion compensation as well as the general operating conditions employed.

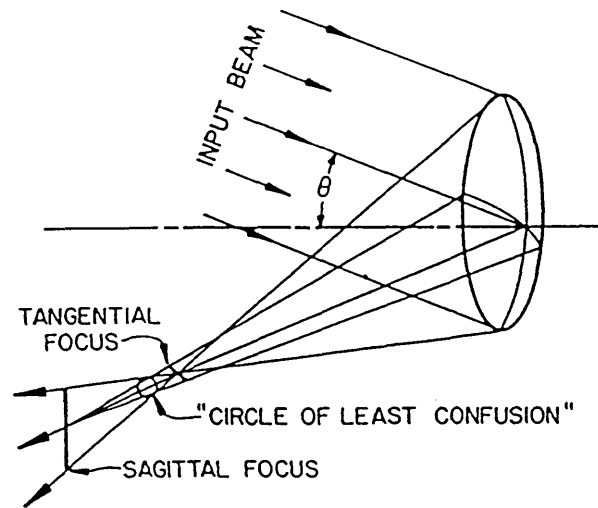
3.2.1 Coumarin 102 active gain jet section

The C102 active gain section is illustrated in Fig. 3.3. The C102 jet nozzle was 700 μm thick, giving a jet thickness of 400 μm in the lasing area. This rather thick jet was used to increase the absorption of the pump beam since the absorption cross section of C102 at $\sim 360 \text{ nm}$ is relatively low, only $4.6 \times 10^{-17} \text{ cm}^2$, approximately

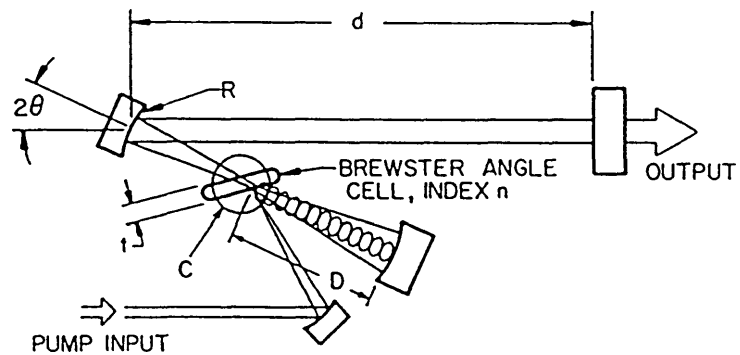
one third of the value for Rhodamine 6G at 500 nm. Thus it is important to optimise the jet thickness and the C102 concentration to use as much pump radiation as possible. Using a typical 3 mirror and 4 mirror cavity, with a 400 μm thick jet, C102 at $3.9 \times 10^{-3} \text{ M}$ concentration was found to give the best slope efficiency. That is, with 1.5 gram of Coumarin powder first dissolved in 300 cc of Benzyl Alcohol and 1.2 L of Ethylene Glycol, 88 percent of the pump radiation was absorbed, better than the typical value of 80 percent. The minimum pumping threshold in this case is only 110 mW, much lower than the 400 mW previously reported^[3,3].

In the 3 mirror configuration the spherical mirror M_3 used off-axis as the internal folding mirror generates an astigmatic focus as show in Fig. 3.5. The rays lying in the incidence plane see the mirror's curvature foreshortened into a tighter arc and come to an out of plane line focus, the tangential focus, ahead of the on axis focal length; and the rays lying in a plane orthogonal to the incidence plane see not foreshortening and reach a line focus, the sagittal focus lying in the incidence plane beyond the on axis focal length. In between the line foci, the beam shape becomes round, the "circle of least confusion" but at a considerably larger focal area. Fortunately, an astigmatism of the opposite sense results when a flat plate is placed in the focused beam in the same plane of incidence but at an angle off normal incidence. In the dye laser this will be the dye cell or jet stream. It can compensate the fold mirror astigmatism and restore the small round focus. The incidence angle for the dye jet is Brewster's angle θ_B , where $\tan \theta_B = n$, the index of refraction of the plane, for minimum resonator losses. For a dye jet of thickness t , the correct fold angle 2θ between the collimated beam and the focused beam that achieves compensation is

$$R \sin \theta \tan \theta = 2t \frac{(n^2 - 1) \sqrt{n^2 + 1}}{n^4} \quad (3.1)$$



(a)



(b)

Fig. 3.5 (a) Astigmatism of an off-axis spherical mirror, (b) its compensation by a Brewster angle dye jet plate.

The free jet of dye dissolved in a viscous solvent like ethylene glycol, squirted at 10 msec^{-1} from a nozzle with a slotted tip and caught in an open tube returning the solution to a fluid pump gives adequate surface flatness and good stability. Dye jets are more economical than optical quality cells and avoid the burning problem of the tightly focusing spot on the dye cells. Astigmatism can also be reduced by closing up the fold angle, since the jet dimension is only 5 mm wide by 0.4 mm thick, the compensation is more effective.

When the active jet folding section goes into the four mirrors configuration as shown in Fig. 3.3, the symmetry forces the major and minor axes of the elliptical beam waist to be simultaneously minimum at the same spatial position. They are nearly equal for this condition so that the waist is circular. A maximum degree of focusing of the pump beam and laser radiation within the dye jet leads to the lowest oscillation threshold. Indeed, we have experimentally achieved a minimum threshold of 110 mW, with improved transverse mode structure and enhanced overall stability and reproducibility.

3.2.2 DOCI passive saturable absorber jet section

The saturable absorber jet section of the CPM uses the "crossed" arrangement as shown in Fig. 3.4. It has the further advantage of reducing cavity losses due to coma. The coma of one of the focusing mirrors M_5 produces a field distribution in the focal plane that is asymmetric with respect to the optical axis. In this "crossed" arrangement the directions of asymmetry of both mirrors M_4 and M_5 coincide so that

the comatic field distribution of the focal plane is re-imaged by the second mirror M_4 into a field distribution that is closer to a Gaussian than in the " Z " folding arrangement, thus reducing the overall loss of the cavity^[3.5].

The saturable absorber jet fold is more tightly focused than the gain jet. The radius of curvatures of M_4 and M_5 are both 25 mm, while in the gain fold the radius of curvatures of M_2 and M_3 are both 50 mm. The saturable absorber jet is thinner than the active jet, typically tens of μm . This is to optimise the colliding pulses interaction. The saturable absorber nozzle is also narrower which makes it easier to fine adjust the focusing of the SA jet for stable operation and for generating the shortest pulse duration ^[3.6].

The colliding pulse interaction in the saturable jet involves a grating buildup as the two counter-propagating pulses overlap^[3.7]. The influence of coherent coupling in colliding pulse systems is essentially equivalent to that of an increased saturable absorber cross section. Compared to other passive methods, considerably shorter pulses with a more favorable stability region can be obtained^[3.8]. In addition, by using gain and saturable absorber jets separated by a quarter of the ring cavity length, the pulses are timed so that they arrive at the active medium equally spaced in time and thus both pulses will see the same maximum saturable gain.

Another variation on the CPM ring cavity is the antiresonant ring^[3.9] which gives similar results but is more critical in alignment.

3.2.3 Dispersion compensation

Pulse durations from passively mode-locked dye lasers have decreased by almost three orders of magnitude since 1972, yet the laser dye combination has remained essentially the same with the gain dye being derived from the rhodamine family and the absorber dye from the cyanine family. Thus, the reductions in pulse durations have been principally due to improvements in laser theory and resonator design as opposed to the development of new lasing media. A significant step forward in generating femtosecond pulses was made with the introduction of the colliding pulse mode-locked (CPM) ring laser resonator which for the first time generated pulses of less than 100 fs duration. Since then, many ring cavity designs have been studied both experimentally and theoretically^{[3.10]–[3.20]}. These studies have revealed that for pulse durations under 100 fs, the effects of intracavity group velocity dispersion (GVD) and self phase modulation (SPM) become increasingly important. It is clear that proper balancing of SPM and GVD could yield soliton-like pulse shaping and reduced pulse durations.

The effects of dispersion in the laser cavity will : (1) increase the width of the output pulse τ_{out} compared to that of the input pulse by a factor

$$\frac{\tau_{out}}{\tau_{in}} = (1 + \frac{\phi''^2}{4\beta^2}) \quad (3.2)$$

where $\beta = \tau_{in}^2 / (8 \ln 2)$, and (2) produce a frequency sweep in the output pulse with the sign opposite to that of ϕ''^2 ^[3.22].

The role of cavity dispersion in CW mode-locked lasers can be classified as [A].

normal dispersion in transparent materials such as blocks of glass or quartz and dye jet streams; [B]. anomalous dispersion of the absorber and amplifier; [C]. self phase modulation within the saturable absorber, and [D]. the dispersion of dielectric coated mirrors.

[A]. Normal dispersion in transparent materials

The phase shift of an EM wave of the form $e^{i(\omega t - kz)}$ after traversing an effective length L_e of a transparent material is

$$\phi(\omega) = -\omega n(\omega) \frac{L_e}{C_o} \quad (3.3)$$

where $n(\omega)$ is the refractive index of the material, $L_e = \frac{L}{\cos \theta_r}$, L is the actual material length and θ_r is the refractive angle, and C_o is the velocity of light in the vacuum. The second derivative is

$$\phi'' = -\frac{L_e}{C_o} \left(2 \frac{dn}{d\omega} + \omega \frac{d^2 n}{d\omega^2} \right) \quad (3.4)$$

To calculate ϕ'' we need to know the variation of the refractive index with frequency ω or with wavelength λ . ϕ'' can also be expressed as

$$\phi'' = -\frac{L_e \lambda^3}{2\pi C_o^2} \frac{d^2 n}{d\lambda^2} \quad (3.5)$$

The behaviour of the refractive index of ethylene glycol as a function of wavelength has been calculated by fitting the experimental values by means of the Sellmeier

equation^{[3.21][3.22]}. For a typical ethylene glycol jet of 100 μm thickness and inclined at Brewster angle we thus get^[3.22] $\phi'' = -8.4 \times 10^{-30} \text{s}^2$.

The dispersion equation of optical quality quartz is^[3.23]

$$n^2 - 1 = \frac{0.6961663\lambda^2}{\lambda^2 - (0.0684043)^2} + \frac{0.4079426\lambda^2}{\lambda^2 - (0.1162414)^2} + \frac{0.8974794\lambda^2}{\lambda^2 - (0.9.896161)^2} \quad (3.6)$$

where λ is in μm . At $\lambda = 0.5\mu\text{m}$, $\frac{d^2n}{d\lambda^2} = 0.4788\mu\text{m}^{-2}$, or $4.788 \times 10^{11} \text{m}^{-2}$, and for 1 mm thick quartz with $n = 1.462$, at the Brewster angle $\theta_B = 55.6^\circ$, the effective length $L_e = \frac{L}{\cos\theta_B}$ is 1.77 mm. Thus for 1 mm effective length we get $\phi'' = -191 \times 10^{-30} \text{s}^2$. The value of ϕ'' for flint F_2 glass has also been calculated and is about three times that of quartz. We can see that the dispersion due to these transparent materials is normal (positive) dispersion and always produces a positive frequency chirp.

[B]. Anomalous dispersion of absorber and amplifier

For the anomalous dispersion arising from the unsaturated loss or gain of the absorber or amplifier, consider a transition centred at frequency ω_0 and with Lorentzian lineshape. The corresponding refractive index n at frequency ω is

$$n = n_0 + \frac{\pi}{2}(\omega_0 - \omega)\left(\frac{c_0}{\omega_0}\right)g(\omega_0 - \omega)\alpha \quad (3.7)$$

where n_0 is the refractive index of the liquid, such as ethylene glycol in which the absorber or amplifier dye is dissolved; α is the absorption (positive) or gain (negative) coefficient at the peak of the transition; and $g(\omega_0 - \omega)$ is the Lorentzian lineshape

function given by

$$g = \frac{2}{\pi \Delta \omega_0} \left(1 + \left[\frac{2(\omega_0 - \omega)^2}{\Delta \omega_0} \right] \right) \quad (3.8)$$

$\Delta \omega_0$ is the width (FWHM) of the lineshape.

Svelto^[3.22] has calculated ϕ'' by inserting equation (3.7) into equation (3.4) and has found that within the usual mode-locked region only the saturable absorber and its photoisomer make appreciable contributions to the dispersion. These two contributions are almost always opposite in sign and tend to cancel. The extent to which this occurs is however difficult to assess since the amount of saturable absorber converted into the photoisomer during the mode-locking operation needs to be investigated more precisely.

[C]. Self phase modulation within the saturable absorber

When a pulse of sufficient intensity passes through a saturable absorber ~~that is tightly focused~~, its absorption coefficient α changes in time due to the absorber saturation. This results in a time change of the refractive index $n(t)$ which in turn induces a time change of the phase of the form

$$\phi(t) = -\omega n(t) \frac{L_e}{C_o} \quad (3.9)$$

This self phase modulation phenomenon has been used to explain the chirp observed in mode-locked dye lasers^{[3.8][3.24]}.

The absorption coefficient α of the saturable absorber can be written as

$$\alpha(t) = \alpha_0 e^{-[\frac{\Gamma(t)}{\Gamma_s}]} \quad (3.10)$$

where α_0 is the unsaturated absorption coefficient, $\Gamma(t)$ is the laser energy fluence at time t , $\Gamma(t) = \int_{-\infty}^t I(t) dt$, $I(t)$ is the laser intensity, Γ_s is the saturation fluence of the saturable absorber, $\Gamma_s = \frac{h\nu}{\sigma}$, and σ is the saturable absorber cross section at the laser wavelength.

So $\phi(t)$ can be expressed as

$$\phi(t) = \phi_0 - \frac{\pi}{2}(\omega_0 - \omega_L)g(\omega_0 - \omega_L)L_e\alpha(t) \quad (3.11)$$

where ω_L is the central frequency of the laser pulse and $\phi_0 = -\omega_0 n_0 \frac{L_e}{c_0}$ is the phase shift of a wave travelling through a length L_e in the ethylene glycol jet stream. For two counter-propagating pulses in the CPM regime, the variation in angular frequency $\delta\omega$ during the pulse duration τ (FWHM) and for an intensity pulshape of sech^2 is^[3.22]

$$\delta\omega = \frac{d\phi}{dt} = -1.56\pi(\omega_0 - \omega_L)\frac{g(\omega_0 - \omega_L)}{\tau}L_e\alpha_0\frac{\Gamma_t^2}{\Gamma_s} \quad (3.12)$$

From equation (3.11) and equation (3.12) we can see that for $\omega_L = \omega_0$ no SPM is produced within the saturable absorber. This is clearly the case since at resonance the absorption lineshape does not contribute to the refractive index of the material as shown from equation (3.7).

For the typical CPM regime with $\tau = 100fs$, $(\omega_0 - \omega_L) = 1.64 \times 10^{14} s^{-1}$, $\Delta\omega_0 = 2.37 \times 10^{14} s^{-1}$, $\alpha_0 L_0 = 0.4$, and assuming $(\frac{\Gamma_t}{\Gamma_s}) = 0.3$, the resulting value for $\delta\omega$ is $\delta\omega = -2.7 \times 10^{11} s^{-1}$. For other situation with higher value of $\frac{\Gamma_t}{\Gamma_s}$ equals to $0.6 \sim 3$, $\delta\omega$ could be equal to $-24 \times 10^{11} s^{-1}$ [3.22].

For comparison with the effects of dispersion considered before, we assume that the pulse after passing through the saturable absorber still has the same shape and duration, but has acquired a linear frequency chirp $\delta\omega$. From the Fourier transform of this pulse we get the phase change $\phi(\omega)$ in the frequency domain, from which we can calculate the quantity $\phi''(\omega_L)$ at the laser frequency. The values thus obtained are $\phi'' = 36 \times 10^{-30} s^2$ and $\phi'' = 300 \times 10^{-30} s^2$ corresponding to $\delta\omega = -2.7 \times 10^{11} s^{-1}$ and $\delta\omega = -24 \times 10^{11} s^{-1}$ respectively.

The SPM in the amplifier dye jet arising from the gain saturation can be ignored because of the less tight focusing which gives greater spot size to the beam and therefore lower saturation level, and also because of the absence of the grating formation of the colliding pulses which only occurs in the saturable absorber jet.

[D]. The dispersion of dielectric coated mirrors

For multilayer dielectric mirrors the reflected wave is produced by the interference of the multiple reflections that take place within the structure. This will produce a phase shift $\phi(\omega)$ of the reflected wave which depends on the laser frequency. Thus a dielectric mirror acts as a source of GVD whose dispersive power is given by $\phi''(\omega)$.

The general method of calculating the electric field reflection coefficient r of

a multilayer is based on a matrix formulation of boundary conditions at the layer surfaces derived from Maxwell's equations^[3.22]. The intensity reflectance $R = |r|^2$, the phase shift $\phi(\omega)$ and the second derivative $\phi'' = \frac{d^2\phi}{d(\frac{\omega}{\omega_m})}$ can be evaluated as a function of $\frac{\omega}{\omega_m}$, where ω_m is the central frequency of the mirror coating and $\phi'' = \frac{d^2\phi}{d\omega^2}$ is related to ϕ'' by $\phi'' = \frac{\phi''}{\omega_m^2}$.

We can see that ϕ'' is zero for $\omega = \omega_m$, and it increases rapidly away from the mirror resonance. The sign of ϕ'' is either positive or negative depending on whether the mirror is used toward the red-shifted or the blue-shifted sides of the reflectivity curve. Thus if the dielectric mirror is used exactly at resonance, it does not contribute to cavity dispersion. However, if the mirror is used at one of the two edges of the reflectivity curve, as the laser output is tuned to different wavelengths, high values of ϕ'' will occur. The ultrashort pulse bandwidth and wavelength detuning the dielectric coated mirror typically gives $\phi'' = 200 \times 10^{-30} \text{ s}^2$ for the cavity dispersion.

Table 3.1 summarizes the cavity dispersion of CPM dye lasers. We see that the largest contributions to dispersion arise from the transparent materials such as glass or quartz, and the multilayer dielectric mirrors. The SPM in the saturable absorber also results in a comparable value of ϕ'' , but its sign is opposite to that of the transparent materials. The chirp arising from SPM can therefore be offset by a suitable length of transparent materials and this leads to pulse compression.

Table 3.1 Dispersion contributions from cavity elements

Cavity components	Dispersion contributions		Sign of GVD	Frequency chirp	Dispersion Parameter
	$\frac{d^2\phi}{d\omega^2}$	$(10^{-30} s^2)$			
Ethylene glycol jet stream (100 μm)	- 8.4		+	+	-
Quartz (1 mm)	-191		+	+	-
Flint glass (1 mm)	-550		+	+	-
Anomalous dispersion in DOCl	7.5		-	-	+
Anomalous dispersion in DOCl photoisomer	- 32		+	+	-
Dielectric coated mirror	a few hundred + or -		- or +	- or +	+ or -
Self phase modulation in DOCl	30 to 300		-	-	
Prisms pair (25 mm)	300		-	-	+
Gratings pair (1 m)	124000		-	-	+

A numerical estimation of the effect of dispersion on the pulse duration can be obtained by using the arguments of Stix and Ippen^[3.7]. For a dispersionless cavity, the laser pulsewidth can be obtained by equating the pulse-shortening velocity ς due to the saturation of both of the absorber and amplifier media, to the per-pass spreading due to the band filtering elements. For a Gaussian pulse of pulsewidth τ , passing through a Gaussian filter whose transmission to the electric field of the pulse is $e^{-(\frac{\omega}{\omega_c})^2}$, the spreading per pass due to the filter is $(\frac{\delta\tau}{\tau})_f = \frac{4 \ln 2}{\omega_c^2 \tau_1^2}$. We can write

$$\frac{4 \ln 2}{\omega_c^2 \tau_1^2} = \varsigma \quad (3.13)$$

where τ_1 is the steady state pulse duration.

Now consider a dispersive cavity, for a Gaussian pulsewidth τ passing through the dispersive element characterized by its second derivative ϕ'' . The spreading per pass from equation (3.2) is $(\frac{\delta\tau}{\tau})_d = 8 \ln^2 2 \frac{\phi''^2}{\tau^4}$. The new steady state pulse duration τ_2 in the presence of dispersion is

$$\frac{4 \ln 2}{\omega_c^2 \tau_2^2} + (8 \ln^2 2) \frac{\phi''^2}{\tau_2^4} = \varsigma \quad (3.14)$$

Equation (3.13) and equation (3.14) show that for the same values of ς and ω_c , the pulsewidth of a dispersive cavity is expected to be larger than that of a cavity without dispersion.

If we assume that the steady-state pulsewidth in a dispersionless cavity is $\tau_1 = 60 \text{ fs}$ and $\varsigma = 1 \times 10^{-2}$, the pulsewidth in the presence of dispersion given

by $\phi'' = 200 \times 10^{-30} \text{s}^2$, will be $\tau_2 = 80 \text{fs}$. However, for the case of $\tau_1 = 150 \text{fs}$, and for the same value of ω_c , the pulse broadening due to the dispersion contribution would be much less significant. This is because the broadening due to the dispersion which is proportional to τ^{-4} , is much more strongly dependent on the pulsewidth than the broadening due to the bandwidth-limiting elements which is proportional to τ^{-2} . The dispersion effects should have significant influence on the mode-locking behaviour for pulsewidth below 100 fs.

Indeed, we can see this from the definition of the dispersion parameter D defined as^[3.25]

$$D(\lambda) = \frac{1}{L} \frac{d\tau}{d\lambda} \quad (3.15)$$

which is normally expressed in units of picoseconds of temporal spread, per nanometre of spectral bandwidth per kilometre of distance propagated in the dispersive medium. Because of its dependence on spectral bandwidth, GVD can be significant for an ultrashort pulse (very wide band) passing through only millimetres of glass. The value of L for which GVD roughly doubles the width of an incident pulse is a convenient characteristic of the process and is given by

$$L_{disp} = \frac{c\Delta t^2}{k\lambda^2 |D|} \quad (3.16)$$

where Δt is the pulse duration and k is a parameter that depends on the pulse shape. For a 1 ps Gaussian pulse, for which $K = 0.441$, $L_{disp} = 6 \text{m}$ in fused silica.

However, the presence of Δt^2 in the numerator of the formula means that for a 10 fs pulse L_{disp} is as small as 0.6 mm.

Avramopoulos and New^[3.26] have modeled the influence of phase effects on the pulse formation in a passive mode-locked laser taking into account the nonlinear saturation of the gain and absorption media, linear loss, bandwidth limitation, noise, group velocity dispersion as well as phase modulation and have given a clear insight into this process.

From table 3.1 we can see that the overall dispersion contributions in the CPM dye laser could be either positive or negative and could have a wide range and values depending on the choice of the cavity components and the operating conditions. Unless special precautions are taken to compensate for the frequency chirp arising from different cavity elements ultrashort pulse generation is difficult to achieve. The CPM set up in our system employs a four-prism sequence as shown in Fig. 3.6 that can introduce dispersion of either sign to balance the intracavity self phase modulation, group velocity dispersion, saturable absorption and saturable gain to generate ultrashort pulses.

We can use the dispersion parameter as defined by equation (3.15) to calculate the dispersions of this prism quadruplet. Let $P(\omega)$ be the optical path length, $P = \int n(x)dx$, of the mode axis in a resonator at frequency ω . The time taken by a light pulse to traverse this path is $\frac{P}{c}$ and

$$\tau = \frac{d(2\pi\frac{P}{\lambda})}{d\omega} = \frac{1}{c}(P - \lambda\frac{dP}{d\lambda}) \quad (3.17)$$

where λ is the free space wavelength, $\lambda = \frac{2\pi c}{\omega}$, $\omega = 2\pi\nu$. The group velocity dispersion parameter D is then given by

$$D(\lambda) = \frac{1}{L} \frac{d\tau}{d\lambda} = -\left(\frac{\lambda}{cL}\right) \frac{d^2 P}{d\lambda^2} \quad (3.15)$$

where L is the physical length of the light path and P is the optical path length. To obtain $\frac{d^2 P}{d\lambda^2}$, consider the rays that propagate near the apices of the prisms. Let the extreme ray that passes from apex to apex be a reference ray, and define its slant length between prisms 1 and 2 as ℓ .

Using the optical construction in Fig. 3.7, the optical path length of a ray CDE that propagates at an angle with respect to the reference ray can be calculated.

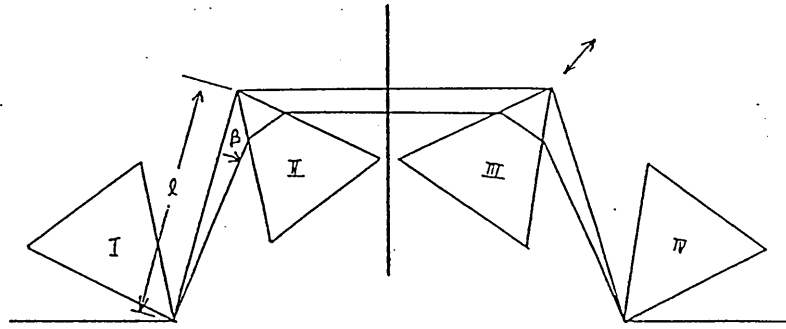


Fig. 3.6 Four prism sequence for intracavity dispersion compensation.

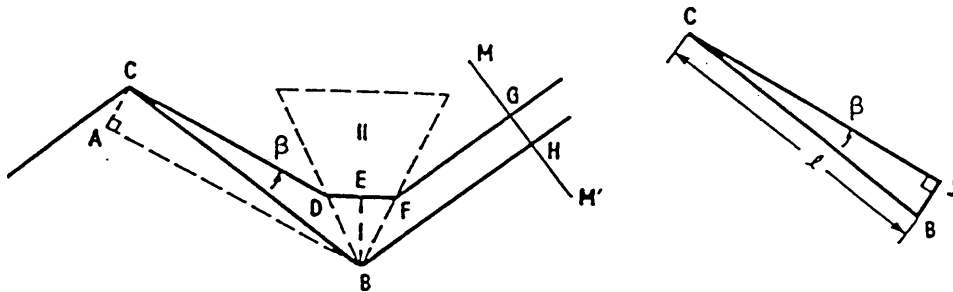


Fig. 3.7 Construction for calculating the paths CDE, EFG, and BH.

Path CDE and path AB are equal, because both AC and BE are possible wavefronts, and path CJ is equal to path AB by construction. So the optical path length P of CDE is

$$P = L \cos \beta \quad (3.18)$$

and since the optical path length EFG and BH are equal because both BE and GH are both possible wavefronts. Optical path FG and BH are both parallel to the incident ray, and hence to each other because the initial prism pair is equivalent in its effect on the ray direction to a slab having parallel faces. The section of path indicated by rays EFG and BH makes no contribution to $\frac{d^2 P}{d\lambda^2}$.

By symmetry, the second pair of prisms introduces the same amount of dispersion as the first pair but reverses the transverse displacement of the rays. The final rays are therefore collinear with each other and collinear with the path of the incident rays.

The total optical path that contributes to the dispersion is therefore

$$P = 2L \cos \beta \quad (3.19)$$

and

$$\frac{dP}{d\beta} = -2L \sin \beta \quad (3.20)$$

$$\frac{d^2 P}{d\beta^2} = -2L \cos \beta \quad (3.21)$$

We need the second derivative of P with respect to λ . The angle β is a function of the prism index n , which is in turn a function of the wavelength λ . Since n is a function of λ , we can express the derivatives of P with respect to λ in terms of derivatives of P with respect to n by

$$\frac{dP}{d\lambda} = \frac{dn}{d\lambda} \frac{dp}{dn} \quad (3.22)$$

$$\frac{d^2 P}{d\lambda^2} = \frac{d^2 n}{d\lambda^2} \frac{dp}{dn} + \left(\frac{dn}{d\lambda}\right)^2 \frac{d^2 p}{dn^2} \quad (3.23)$$

and since n is a function of β , we can express the derivatives with respect to n as derivatives with respect to β employing the chain rule for derivatives as

$$\frac{d^2 P}{d\lambda^2} = \left[\frac{d^2 n}{d\lambda^2} \frac{d\beta}{dn} + \left(\frac{dn}{d\lambda}\right)^2 \frac{d^2 \beta}{dn^2} \right] \frac{dP}{d\beta} + \left(\frac{dn}{d\lambda}\right)^2 \frac{d\beta^2}{dn} \frac{d^2 P}{d\beta^2} \quad (3.24)$$

Using the geometry of a light ray traversing a prism at minimum deviation^[3.27], the angle of incidence of the beam at prism 1, ϕ_1 is fixed, but the angle that the transmitted beam makes with the normal to the prism face ϕ_2 is variable accordingly. The respective interior angles ϕ_1' and ϕ_2' are given by $n \sin \phi_1' = 1 \sin \phi_1$, and $n \sin \phi_2' = 1 \sin \phi_2$. For prism index n and apex angle α , Snell's law and the relation $\alpha = \phi_1' + \phi_2'$ gives

$$\frac{d\phi_2}{dn} = \frac{[\sin(\phi_2') + \cos(\phi_2')\tan(\phi_1')]}{\cos(\phi_2)} \quad (3.25)$$

$$\frac{d^2\phi_2}{dn^2} = \tan\phi_2\left(\frac{d\phi_2}{dn}\right)^2 - \frac{\tan(\phi_1')}{n}\left(\frac{d\phi_2}{dn}\right) \quad (3.26)$$

For minimum deviation and Brewster angle incidence, $\phi_1' = \phi_2'$, and $\tan\phi_2 = n$. Using $\frac{d\beta}{dn} = -\frac{d\phi_2}{dn}$, $\frac{d^2\beta}{dn^2} = -\frac{d^2\phi_2}{dn^2}$, we can have

$$\frac{d\beta}{dn} = -2 \quad (3.27)$$

$$\frac{d^2\beta}{dn^2} = -4n + \frac{2}{n^3} \quad (3.28)$$

Inserting these relations and equations (3.20) and (3.21) into equation (3.24) we get

$$\frac{d^2P}{d\lambda^2} = 4\ell\sin\beta\left[\frac{d^2n}{d\lambda^2} + \left(\frac{dn}{d\lambda}\right)^2\left(2n - \frac{1}{n^3}\right)\right] - 8\ell\cos\beta\left(\frac{dn}{d\lambda}\right)^2 \quad (3.29)$$

Since β is of the order of the angular deviation of the ray bundle, so $\sin\beta \ll \cos\beta$, $\ell\sin\beta$ need only be of the order of twice the spot size, that is $\sim 2mm$; and $\cos\beta$ can be approximated by unity. For the Spectrosil B quartz at 500 nm, $n = 1.462$, $\frac{dn}{d\lambda} = -0.1089\mu m^{-1}$, and $\frac{d^2n}{d\lambda^2} = 0.4788\mu m^{-2}$. So

$$\frac{d^2 P}{d\lambda^2} = 4.077 - \ell(0.0949) \quad (3.30)$$

where P and ℓ are measured in millimeters, and λ in micrometers. For $\ell \geq 43mm$, the four-prism sequence oriented as shown in Fig. 3.6 has negative dispersion. The magnitude of this negative dispersion can be compared with the positive dispersion of quartz by dividing by $\frac{d^2 n}{d\lambda^2}$ given above. Fig 3.8 shows the equivalent quartz length that can be compensated for by the negative dispersion generated by the prism quadruplet. It is also convenient to introduce positive dispersion for any necessary compensation simply by translating any of the prisms in the direction normal to the prism base line so that the cavity beam can have more glass path and yet the resonator alignment remains the same. The prism quadruplet has been used widely as an intracavity elements because of the low loss and the collinear property of the input and output beams.

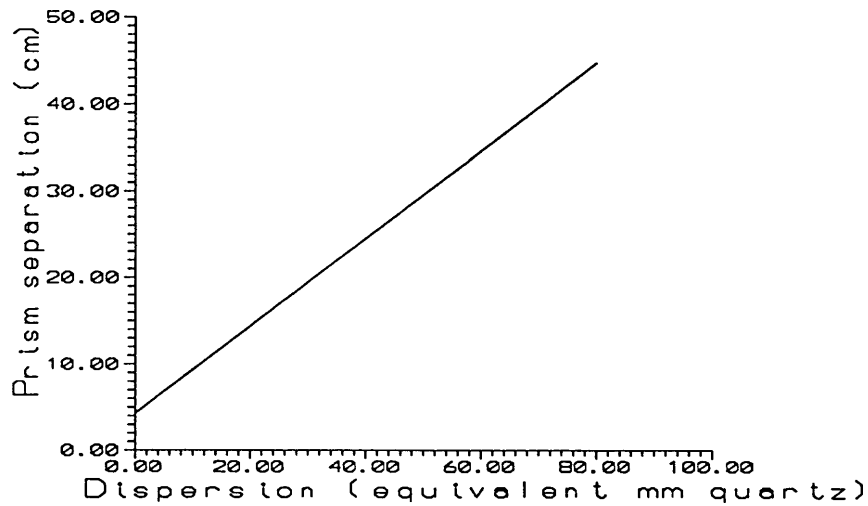


Fig. 3.8 Positive and negative dispersion from the prisms pair.

An alternative component that can produce adjustable negative GVD in the set up is a pair of diffraction gratings^[3.28]. A pair of diffraction gratings can be arranged with their faces and rulings parallel so that the incident laser beam, after having been diffracted twice, will emerge parallel to the input beam. Like the prism pairs, there will be a spatial distribution of the spectral components of the incident beam, and the lower frequency components will traverse a greater distance due to the angular dispersion of the diffraction. The emerging beam can be reflected back through the original grating pair by a mirror in order to remove the spatial distribution of the spectral components by reversing the transverse displacement. The dispersion introduced by a grating pair in the double pass configuration has been derived by Treacy^[3.28] as

$$\phi'' = \frac{d^2\phi}{d\omega^2} = \frac{-8\pi^2 cb}{\omega^3 d^2 [1 - (\frac{2\pi c}{\omega d} - \sin \gamma)^2]} \quad (3.31)$$

where c is the velocity of light in free space, b is the grating separation, ω is the frequency of the incidence beam, d is the grating constant and γ is the angle of incidence. Typically with $d = 1200$ lines per mm, a grating separation of 1 meter, and operating in the visible regime, the corresponding negative GVD is of the order of $10^{-24} s^2$. This method can generate rather high values of negative GVD compared with the prism sequence method. However, the grating pair can not be used inside a dye laser cavity because of their relative high loss. They are invaluable nevertheless, in extracavity applications as fibre optic compressors and chirp pulse amplification systems.

3.2.4 Operating optimisation

The ability to generate the ultrashort pulses has a sensitive dependence on the saturable absorber dye concentration, proper alignment of the cavity elements, properly balanced intracavity group velocity dispersion and self phase modulation, and a suitable balance between the pump intensity and intracavity power.

The laser can be configured to suit different experimental requirements. If we want to increase the output power, we could increase the saturable absorber concentration and accordingly increase the pumping power while still maintaining relatively ultrashort pulse durations but sacrifice the lifetime of the laser dyes, or we could operate the laser at a lower saturable absorber concentration and higher output coupling and sacrifice the shortness of the pulses generated^[3,6]. Typically we are operating the CPM laser at a suitable concentration that gives relatively ultrashort pulses with a tolerance of pump power level variations up to 30 percent above threshold and a quite good slope efficiency.

Adjusting the prisms to introduce excess negative GVD lengthens the pulse duration considerably up to nearly an order of magnitude. The behaviour of the laser depends sensitively on the position of the prisms and the position of the saturable absorber jet relative to the beam waist between the saturable absorber focusing mirror folding. To generate the shortest pulses as the jet is moved closer to focus, it is necessary to translate a prism to introduce more glass path and accordingly less negative GVD. Under these circumstances, it is possible to obtain shorter pulses. Continuing this pattern of adjustment, and monitoring the real time autocorrelation

trace displayed on the oscilloscope, eventually yields the shortest pulses.

For excess negative GVD beyond that which yields the minimum pulse duration, the laser still maintains stable pulse formation but the pulsewidth gradually increases. However, for excess positive GVD, the pulse formation is more unstable and the pulsewidth broadens significantly. This asymmetry in the dependence of the pulsewidth on the sign of the intracavity dispersion has been investigated both experimentally and theoretically. When the intracavity SPM and GVD are properly balanced, a soliton-like shaping mechanism assists the formation of ultrashort stable pulses. However if SPM and GVD are incorrectly balanced, it will hinder the formation of ultrashort stable pulses. The presence of this mechanism manifests itself as an asymmetric behaviour of the pulse duration as a function of the sign of the net cavity GVD^[3,6].

With proper design considerations for the set up and operating characteristics of the passive CPM ring dye laser we can generate sub-hundred fs pulses satisfactorily. Negative intracavity group velocity dispersion is introduced by the prism quadruplet, while positive self phase modulation is generated by the optical Kerr effect in the dye solvents and gain saturation. Under the proper conditions, the operating characteristics involve a novel pulse-shaping mechanism with contributions similar to the cause of soliton formation in optical fibres.

3.3 CPM ring dye laser output results

In a set up configuration as shown in Fig. 3.1, the output of the passive CPM ring dye laser could be tuned to 497.2 nm by choosing proper DOCl saturable absorber concentration to cut the blue spectrum and by using a tuning slit to cut the yellow spectrum. The total output power of the two beams is greater than 10 mW at 4 W input pumping. The mode-locked pulses train runs at 140 MHz repetition rate giving approximately 7 ns separation per pulse. The shortest pulse duration measured is 88 fs. The laser can generate sub-hundred fs pulses satisfactorily.

In order to access the generation of the dispersion compensated ultrashort pulses, the "real-time" autocorrelator was found invaluable. The ultrashort pulse width measurements employ the standard collinear SHG technique using BBO as the frequency doubling crystal. Fig. 3.9 is a schematic of the "real-time" autocorrelator apparatus used. Mirror M_1 is attached to a stepping motor micro-translator^[3.29], used as a reference arm in the real-time mode and as a wide range scanner in the scanning mode. Mirror M_2 is attached to a shaker^[3.30] which moves back and forth with a linear motion as much as 5 mm when driven by a sinusoidal signal. The motion of mirror M_2 scans the pulses from one beam at the SHG crystal through their counterparts, resulting in a repetitive second harmonic signal displayed on an oscilloscope. The output signal from the photomultiplier is added to a square wave which is synchronised with the sinusoidal signal driving the shaker. The fundamental beam is blocked by the narrow band pass filter in front of the photomultiplier. Thus the autocorrelated signal with proper scope internal time base can be displayed in real-

time. This is most convenient when adjusting the dispersion compensation through the saturable absorber jet focusing and the glass path in the prism sequence. The oscilloscope display was calibrated by inserting a glass slide of known thickness into the arm of mirror M_1 and measuring the displacement of the peak of the autocorrelation trace.

Figure 3.10 shows the ultrashort pulses of 88 fs duration measured from the C102/DOCI CPM ring dye laser.

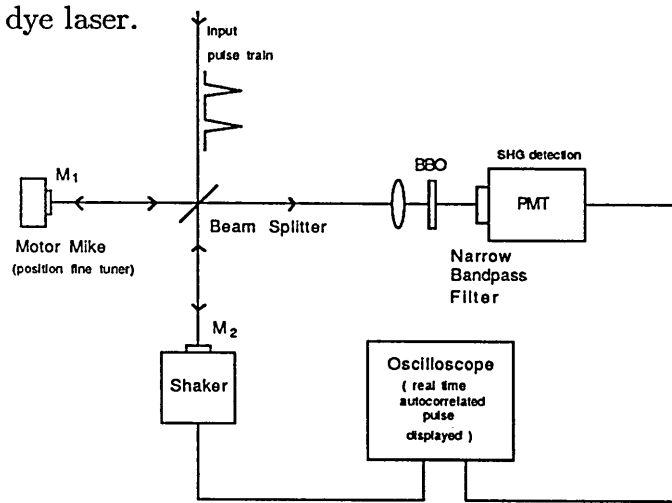


Fig. 3.9 Real time collinear autocorrelator.

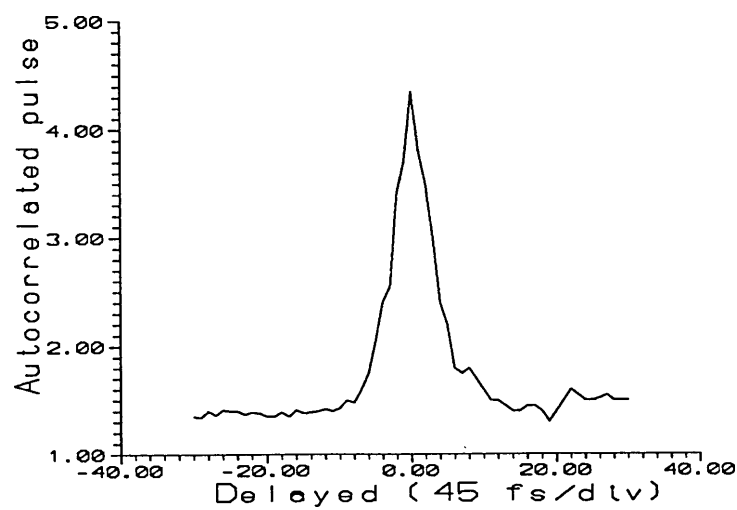


Fig. 3.10 Ultrashort pulses of 88 fs generated from the C102/DOCI CPM ring dye laser.

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Chapter 4

Stable Seed Oscillator

Sub-hundred femtosecond ultrashort pulses from a CPM ring dye laser were generated in the blue green regime, specifically tuned to 497.2 nm, which is exactly double the wavelength of the KrF transition. The oscillator seed pulse was steered through a three stage four pass dye amplifier chain for the preliminary amplification, frequency doubled to 248.6 nm, and amplified again in the KrF excimer amplifier to obtain a high intensity ultrashort UV pulse.

The high intensity laser system requires good stabilities. It is most important to have a stable seed pulse and good synchronisation to both the dye amplifier chain and the KrF amplifier. What is more, the gain bandwidth of the KrF amplifier is typically 2 nm (320 cm^{-1}) FWHM^[4.1] and so it is rather critical to tune the seed oscillator pulses to the exact wavelength with minimum jittering. The amplification produced by the complete system was not totally satisfactory and this was due mainly to the wavelength jittering in the seed oscillator. We had observed from the spectrometer and the wavemeter output that wavelength jitters over ranges that sometimes even exceed the KrF gain bandwidth. Poor amplification and system instability is a serious consequence of this.

The CPM output is very sensitive to the alignment and the operating characteristics^[4.2]. There are many causes that can result in the CPM instabilities, such as:

(1) The commercial Ar^+ pumping source will of course inevitably ~~to~~ have intensity fluctuations even if the light mode is regulated and there will also be a pointing instability^[4.3]. The pumping source could be improved at the manufacture stage with extra cost or by adding ~~a~~ feed back circuitry to minimise the pump source instability^{[4.4][4.5]}. (2) The CPM ring laser has a cavity length greater than 150 cm and so any microscopic unstable movements of the optical mounts could also cause output fluctuations. (3) The active Coumarin 102 jet and the passive DOCI jet are circulating via separate micro-pumps. The vibration of the motor pump sources, as well as the solvent flowing instabilities that varies the jet thickness and the homogeneity will affect the stability of the CPM output. (4) The different methods of wavelength tuning that can be employed also affect the overall output stability.

4.1 Processes of improving seed oscillator stability

It is essential to obtain a seed oscillator output pulse which is as stable as possible. The dominating factors are the isolation and decoupling of the oscillator vibration sources, optimisation of the resonator configuration to generate ultrashort pulses with the best stability, and the choice of wavelength tuning method.

4.1.1 Isolation and decoupling of the circulating vibration sources

The Courmarin 102 dye jet and the DOCI dye jet are both pumped by separate but similar micro-pumps. The motor pump gives huge vibrations that can easily be

felt through the tubing. It is essential to mount each pump on a separate solid base and decouple the vibration to the jet section as much as possible. The tubing has been held with maximum cushion absorption and the nozzle near the jet region is also clamped firmly.

From the interference fringe pattern of the dye jets we were able to see clearly that the dye jets were quite stable and with good homogeneity. The saturable absorber jet is thinner than the active jet, about one fourth that of the active jet, and so a plastic shielding was also applied above the horizontal plane of the beam path to minimise the air turbulence effect in this jet.

4.1.2 Linear cavity

A linear cavity was set up to generate the ultrashort pulses. It is similar to the six mirror cavity described in chapter 3 but with a modified saturable absorber fold section and just one prism pair for the dispersion compensation. Fig. 4.1 shows the configuration of the linear cavity.

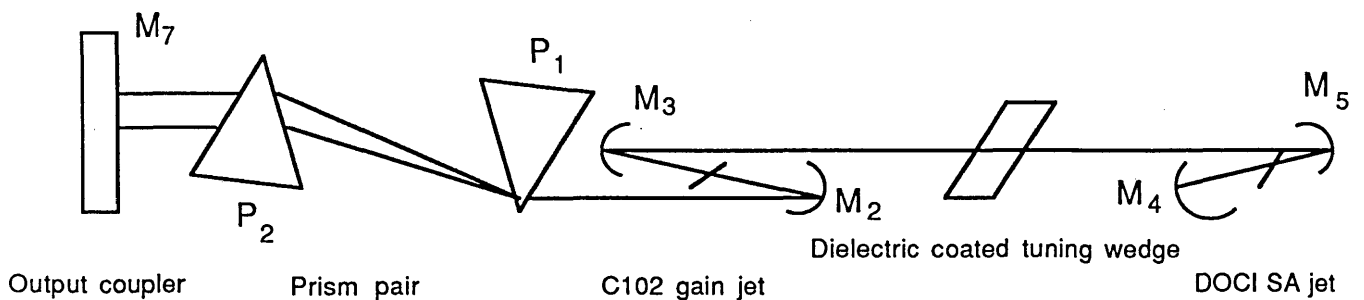


Fig. 4.1 Stable seed laser configuration.

The saturable absorber fold is not of the "cross" type since there is no colliding pulse interaction at that region and due to the physical dimensions of the mirror mounts, it is impossible to align the "cross" fold within the range of the focal length of the mirrors. Just as the saturable absorber cell should be placed near to or directly in contact with the rear mirror of a conventional cavity in order to enhance mode-locking effects, the saturable absorber jet in our cavity was aligned to the focal point of mirror M_5 , and at the centre of the curvature of mirror M_4 . The focal lengths of mirror M_4 and mirror M_5 are the same as in the CPM configuration and are 12.5 mm. The time required for the beam traverse from the saturable absorber jet to the mirror M_4 and back again is 160 ps, still within the relaxation time of the DOCl saturable absorber^[4.6]. Thus the threshold of the mode-locking operation could still be maintained at a low value.

A pair of prisms is aligned to the left hand side of the active jet folding with the total reflecting mirror M_7 at the parallel beam region. This single prism pair configuration can generate the same amount of negative dispersion as the four prism sequence provided the prism separations are the same. The total cavity length had been optimised to give equal spacing for the gain amplification and thus maintain the minimum threshold. Fork et al^[4.7] has also obtained direct generation of 33 fs pulse durations in a linear cavity similar to that of the CPM ring cavity in which they directly generated shortest pulse duration of 27 fs.

This linear cavity has less components compared to the previous CPM configuration with only one prism pair and one reflecting mirror. Thus the linear cavity can

have less optical losses, is easier to align, and has less intracavity dispersion. Yet it still maintains almost all the advantages of the optimised CPM configuration which was described earlier.

4.1.3 Wavelength tuning

One of the most important requirements for generating ultrashort pulses is that within the oscillator cavity there should be no bandwidth limiting element. In the passive mode-locked dye laser bandwidth limitation is most likely to arise from the wavelength tuning elements.

The wavelength tuning mechanism in the passive mode-locked dye laser is achieved by employing the saturable absorber to cut one end of the spectrum and then using a wavelength tuning element to cut the unwanted spectrum or using the tuning element to select the desired wavelength. Typically, the wavelength tuning element is a slit, pinhole, birefringent filter or a dielectric coated tuning wedge.

In our CPM configuration we use a translational slit of variable width in the central region of the prism sequence to select the desired lasing wavelength. It is the easiest and most economic way of tuning, and since there are no bandwidth limiting elements inserted into the cavity, the generation of ultrashort pulse duration will not be limited. However for the wavelength stability considerations this technique is not recommended^[4.8], since any jittered wavelength will still have equal opportunity to lase and fine selection of the lasing wavelength is difficult to obtain. Fork^[4.9]

proposed using curved mirrors instead of the plano mirrors M_6 and M_7 and using a fine pinhole at the focal point in the central region of the prism sequence for wavelength selection. Alternatively, a birefringent filter or a dielectric tuning wedge can be used as tuning elements. The birefringent filter^[4.10] which has been used extensively, consists of multilayers of birefringent crystals at different thicknesses. Once the birefringent filter is tuned, i.e, rotated, it can select the desired lasing wavelength. The dielectric tuning wedge^[4.11] is a thin layer of dielectric coatings in a wedged configuration on a substrate material. As the dielectric wedge is tuned to different thicknesses, it selects the desired lasing wavelength.

Comparison of the four wavelength tuning methods is summarised in Table 4.1. In our linear cavity configuration we use the dielectric tuning wedge for the wavelength selection. It can satisfactorily generate ultrashort pulses with durations around 200 fs which have good wavelength stability. The method has also been reported in the generation of 125 fs pulse durations^[4.11].

4.2 Optimisation of the linear cavity seed oscillator

The linear cavity passively mode-locked dye laser can be used to generate ultrashort pulses. However, the operating characteristics and the intracavity dispersion compensation must be carefully optimised.

Table 4.1 Comparison of wavelength tuning elements

Tuning element	Advantages	Disadvantages
Slit	1. No bandwidth limiting. 2. Inexpensive.	1. Poor wavelength stability.
Pinhole	1. No bandwidth limiting. 2. Improved spectrum shaping and spectrum resolution.	1. No improvement in wavelength stability.
Birefringent filter	1. Self centering of wavelength tuning. 2. Easy to align. 3. Better wavelength stability.	1. Bandwidth limiting element. 2. Introduces cavity losses.
Dielectric tuning wedge	1. Self centering of wavelength tuning. 2. Easy to align. 3. Better wavelength stability. 4. Introduces less cavity losses	1. Bandwidth limiting element. 2. Introduces cavity losses.

The Coumarin 102 and DOCI combination is tunable from 488 nm to 503 nm. For application in our system the first priority is that the seed laser must be tuned to 497.2 nm. This can be achieved by adding the proper saturable absorber concentration and by using the dielectric tuning wedge to tune the wavelength to 497.2 nm. The reason for optimising the saturable absorber concentration is that it ensures better mode-locking depth and the generation of shorter duration pulse as shown in Figure 4.2. Also, as we increase the saturable absorber concentration the output power increases as provided the pumping power ~~can~~ sufficiently exceeds the threshold. Fig. 4.3 shows the experimental results of output power versus saturable absorber concentration. The other aspect in favour of using higher saturable absorber concentration is that it then possesses a higher operating range above threshold. Fig 4.4 shows the slope efficiency of the laser output at two saturable absorber concentrations. At the higher concentration the laser can be pumped almost 48 percent above the threshold, but for the lower saturable absorber concentrations the pumping tolerance is only 10 to 15 percent. Due to the pump power limitation and the practical conditions of keeping the laser lasing for a longer period of time and the ease of restoring the misalignment of the laser it is always advisable to operate the laser at medium to high saturable absorber concentration.

The typical lifetime of the Coumarin 102 active dye in benzyl alcohol and ethylene glycol solution is 40 W-h, and the Ar^+ laser source that is used has a maximum pumping power of 4 W. This implies that under continuous operation the dye can last quite well for the experimental investigations.



Fig. 4.2 Pulsewidth versus DOCl saturable absorber concentrations.

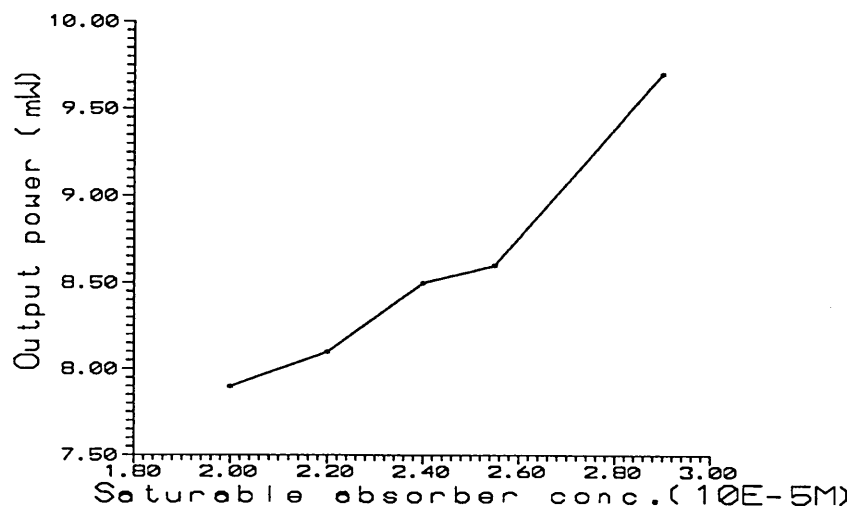


Fig. 4.3 Stable seed laser output power at different S. A. concentrations.

Organic dyes have limited productive lifetime. The factors that limit the lifetime of laser dyes are thought to be the chemical and photochemical degradation of the dye in solution. The lifetime of a dye is often specified in terms of watts-hours. This power-lifetime product is a measure of the pump laser power that has been used to excite the dye. The degradation must be in part due to thermally activated chemistry and /or actual laser induced photochemistry in the dye solution. The chemistry of the dyes can be quite complicated as is evidenced by the fact that the lifetime of the dyes' gain can even be affected by the type of metal plumbing components used in the dye circulating system. The lifetime of the typical cw dye range from ~ 40 W-h (Comarin 480) to several 1000 W-h (Rh 6G).

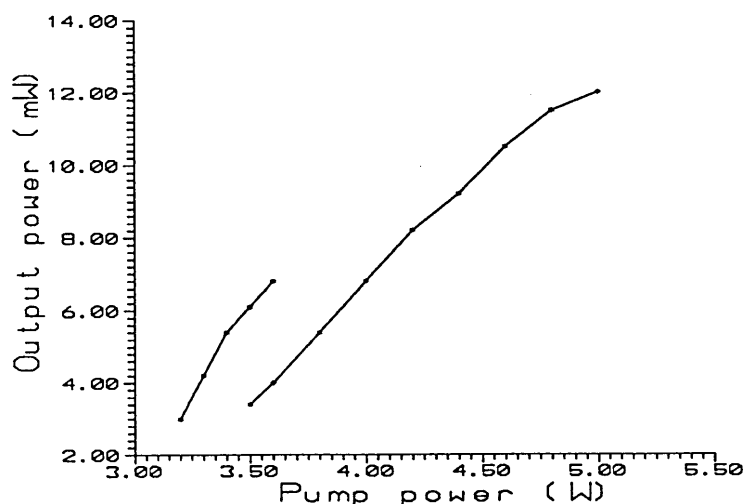


Fig. 4.4 Pumping tolerance at different S. A. concentrations.
wide range shows concentration at 3×10^{-5} M,
short range shows concentration at 1.5×10^{-5} M.

The dyes used in our system, therefore, have a limited lifetime. This is especially true for the Coumarin dye since it is pumped continuously by the high intensity UV laser source. To lengthen the lifetime of the dyes we have used a chopper between the pump source and the seed oscillator during the alignment of the system. In this way the pump source is on for a time period of hundreds of milliseconds when generating the pulse train for the alignment. Once aligned, the seed oscillator is pumped continuously for the characterisation of the system and the experimental investigations. In this way the lifetime of the laser dyes is significantly extended.

To generate the shortest pulse durations it is most important to optimise the intracavity dispersion compensation. A prism pair has been used in this linear cavity configuration to compensate for the dispersion. From the real time display of the autocorrelated pulse on the scope, we can directly see the influence of moving one of the prisms to introduce more or less glass path for the dispersion compensation and in this way choose the path length which leads to the shortest pulse.

It is also rather critical to align the saturable absorber jet position relative to the focal point of the saturable absorber fold in order to operate in the stable regime and with minimum dispersion^[4.2].

Fig. 4.5 shows how the pulsewidth varies as the glass path is altered. The asymmetry results show that it is better to operate the oscillator with a little negative dispersion as its pulsewidth will increase only slightly but not as abruptly as when positive dispersion is introduced. This agrees with previous investigations^{[4.2][4.13][4.14]}.

Once we have tuned the prism to compensate the dispersion and generate the shortest pulse duration, increasing the saturable absorber concentration was found to give no further reduction to the pulse duration. On the other hand, as we go to the more negative or positive dispersion regime, increasing the saturable absorber concentration tends to decrease the pulse duration. This constitutes a three dimensional result for the ultrashort pulse duration versus different prism glass path lengths for the dispersion compensation and saturable absorber concentration as shown in Fig. 4.6.

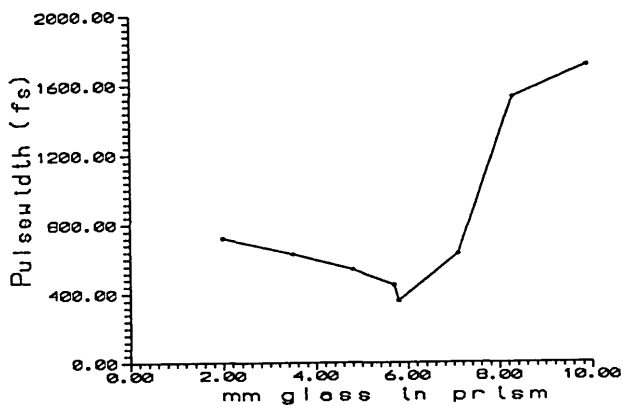


Fig. 4.5 Pulse varies as the glass path length in the prism pair for dispersion compensation is changed.

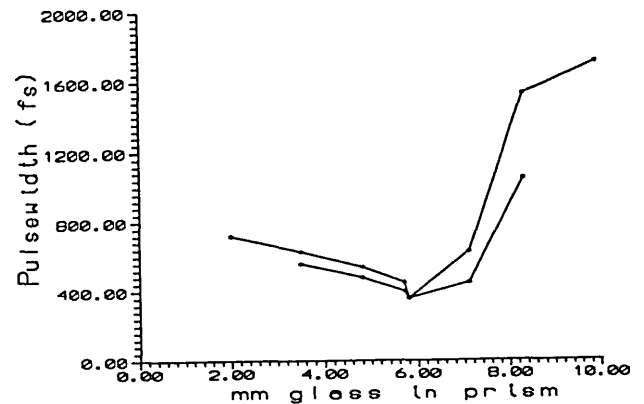


Fig. 4.6 If the glass path length is not optimised to generate the shortest pulse duration, increase saturable absorber concentration will also reduce the pulse duration.

As the laser dyes get old, the shortest pulse duration generated gradually increases. We have measured the shortest pulse duration generated as dyes getting old from 360 fs, $\rightarrow$ 450 fs, $\rightarrow$ 540 fs, $\rightarrow$ 630 fs, and 700 fs, typically increasing from 360 fs to 700 fs over a period of C102/DOCI dyes lifetime.

In this section it has been characterised that with this linear cavity we can generate ultrashort pulses at 497.2 nm, with the output power over 10 mW and pulse durations in the region of 200 fs. The corresponding spectrum measured is shown in Fig. 4.7, and this suggests that the bandwidth is broad enough to support pulses shorter than 150 fs.

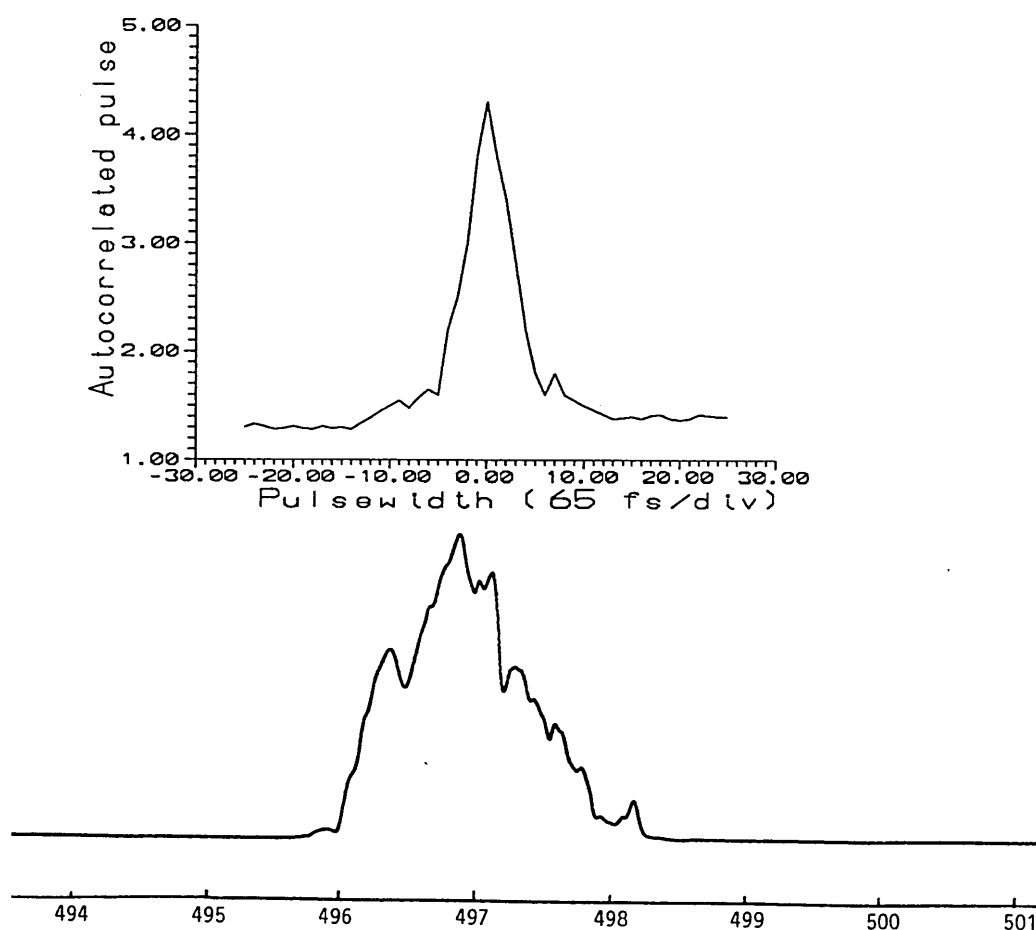


Fig. 4.7 (a) Autocorrelated pulse of the stable seed laser, and (b) the corresponding spectrum.

4.3 Characterisations of wavelength stability

There is still one very important characterisation for the set up of our high intensity ultrashort pulses UV laser system, namely the long term wavelength stability of the seed oscillator.

With proper alignment of the oscillator and suitable operating conditions, the seed oscillator could generate ultrashort pulses with duration of 200 fs, output power greater than 10 mW, at 497.2 nm. However, since there inevitably exist turbulences from the pumping source, jet instabilities, the temperature variations and even mechanical mounting vibrations, the output will suffer intensity fluctuations and wavelength jittering. This needs to be characterised more precisely.

The intensity fluctuations of the seed oscillator can be monitored by the fast response avalanche photodiode and the oscilloscope combination. The maximum intensity fluctuations were found to be less than 5 percent ~~and~~ on a sub-second or longer time scale. This really is not a significant variation since the seed pulse will go through the 3 stage 4 pass dye amplifier and double pass the KrF amplifier in a fraction of this time scale. The complete high intensity UV system shot to shot output intensity variations are indeed dominated by the KrF excimer amplifier fluctuation and the system synchronisation.

Any wavelength jittering, on the other hand, even in the sub-nm ranges, could give rise to a huge system output intensity fluctuation since the gain bandwidth of the KrF amplifier is only 2 nm FWHM. Fig. 4.8 shows the measurement set up to characterise the wavelength jittering of the seed laser oscillator. The output beam

of the seed laser which has both intensity fluctuations and wavelength jitter passes through a 50/50 beam splitter. The first beam is focused onto the fast response silicon avalanche (BPW-28, Teledyne product) photodiode no.1 as a reference signal. The second beam is focused onto a dispersive spectrometer (Hilter monospek 600, dispersion. 13.9 Angstromper mm) and the output from the spectrometer is then focused to another fast response silicon avalanche photodiode no. 2. The output signals from the two photodiodes are then simultaneously displayed on the storage scope and can be recorded to the digitiser.

The spectrum of the seed oscillator was first taken by scanning the spectrometer from 494 nm to 500 nm, and recording the signal output from the silicon photodiode with integrated amplifier on an x-y recorder. The spectrum is shown in Fig. 4.7.

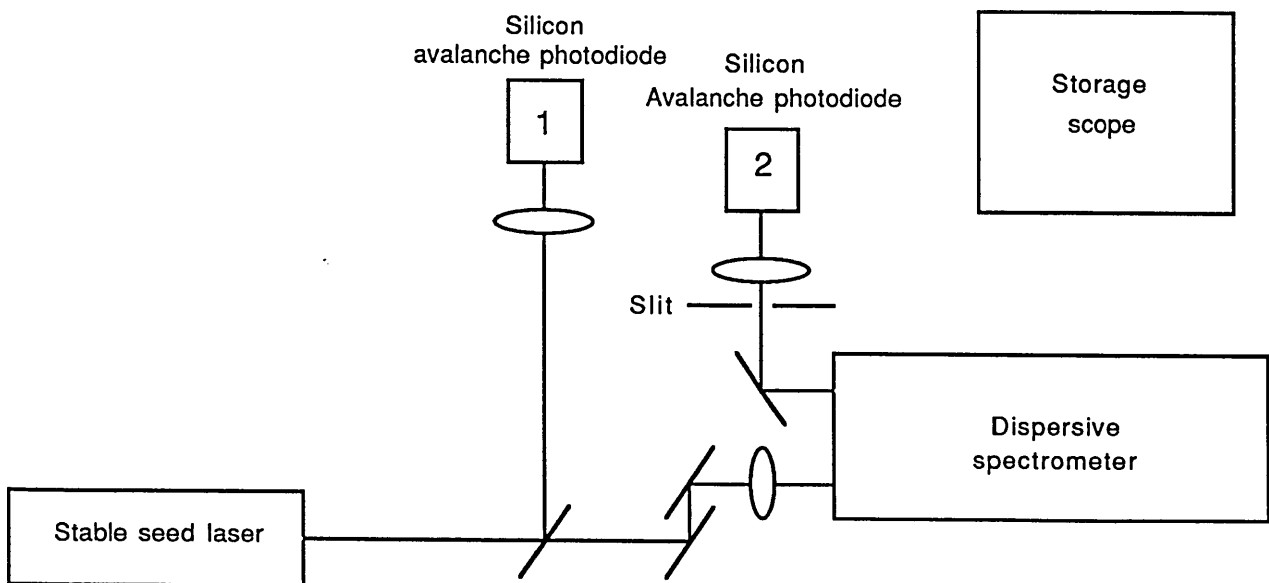


Fig. 4.8 Set up for measuring wavelength jittering.

The display on the x-y recorder gives a maximum spectral width for the pulse of ≈ 2 nm, and from the measurement of the polaroid film the spectral broadening is 1.5 mm, which, since the dispersion calibration of the spectrometer is 1.39 nm per mm, implies that the spectral width is 2.09 nm. The measurement from the film, therefore, agrees well with the direct measurement on the x-y recorder.

If we set up a slit with width smaller than the spectral broadening, and there is still enough signal to be detected by the photodiode, no. 2, then the signal output from the photodiode no. 2 contains not only the intensity fluctuations as seen in photodiode no. 1, but also the dispersive wavelength jittering in it. By taking the ratio of the two signals from 2 to 1, and normalising, the deviation of the values from unity is the wavelength jitter. By varying the time base on the oscilloscope we can investigate the wavelength jitter on various different time scales. We set the total screen time to 2 seconds, and divided the output into 50 equal sections, each representing the average value over 40 ms. The normalised wavelength jitter is shown in Fig. 4.9. This shows only the relative percentage of the wavelength jittering. Since under the same alignment and operating conditions the measured spectra are quite stable, we can use the spectrum shape of Fig. 4.7 to calculate the wavelength jittering as the deviation from the central wavelength in nm. Fig. 4.10 shows the wavelength jittering of the seed laser in nanometer deviation per 40 milliseconds time scale. From the results we can see that the maximum deviation is less than 1 nm, and typically the deviation is less than 0.6 nm, two thirds of the wavelength jittering is less than 0.3 nm. Since the seed laser will be frequency doubled, the deviations of the wavelength jitter is half of the value shown, that is well within the KrF amplifier gain bandwidth.

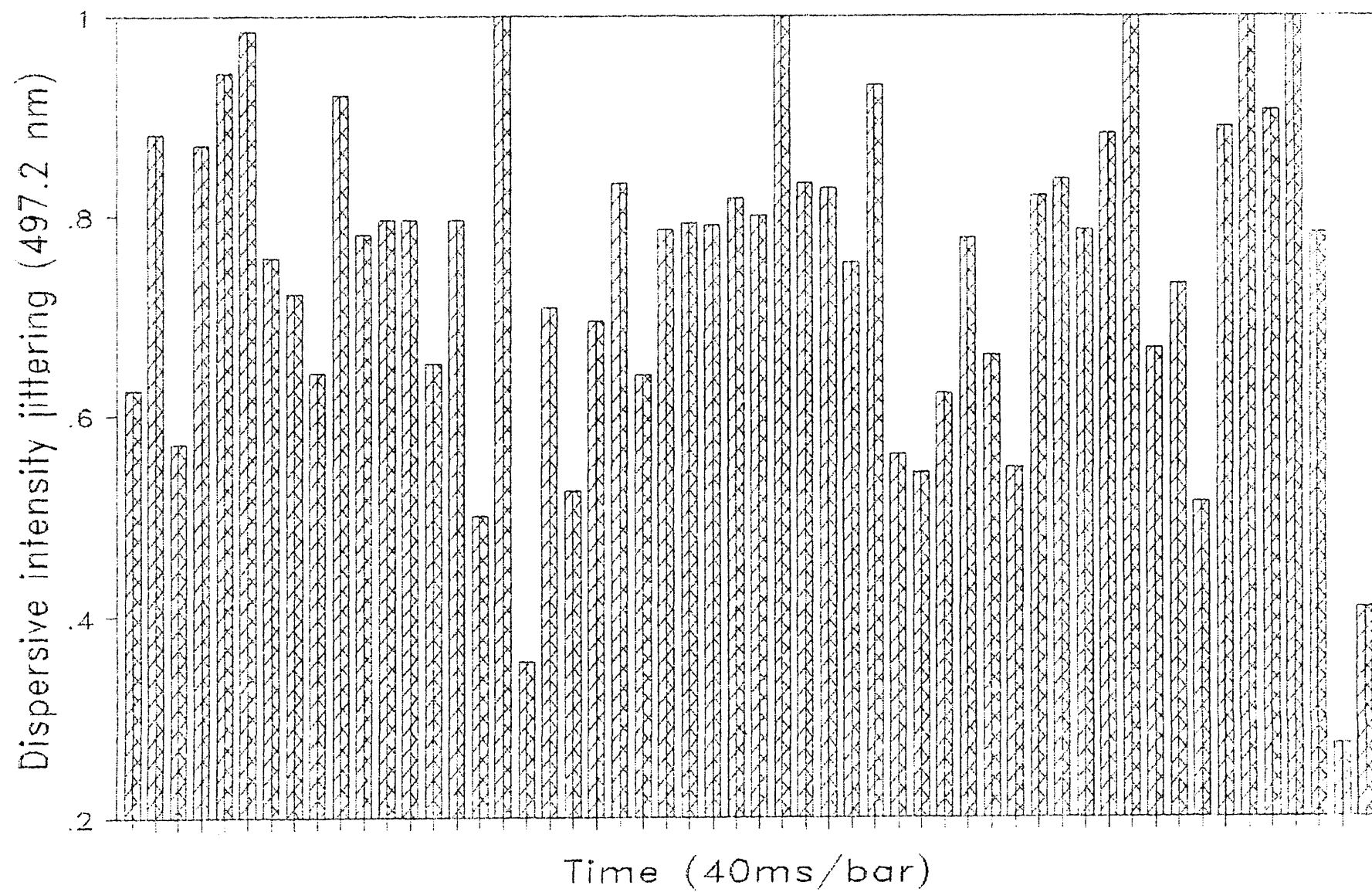


Fig. 4.9 Intensity fluctuation at 497.2 nm dispersive wavelength.

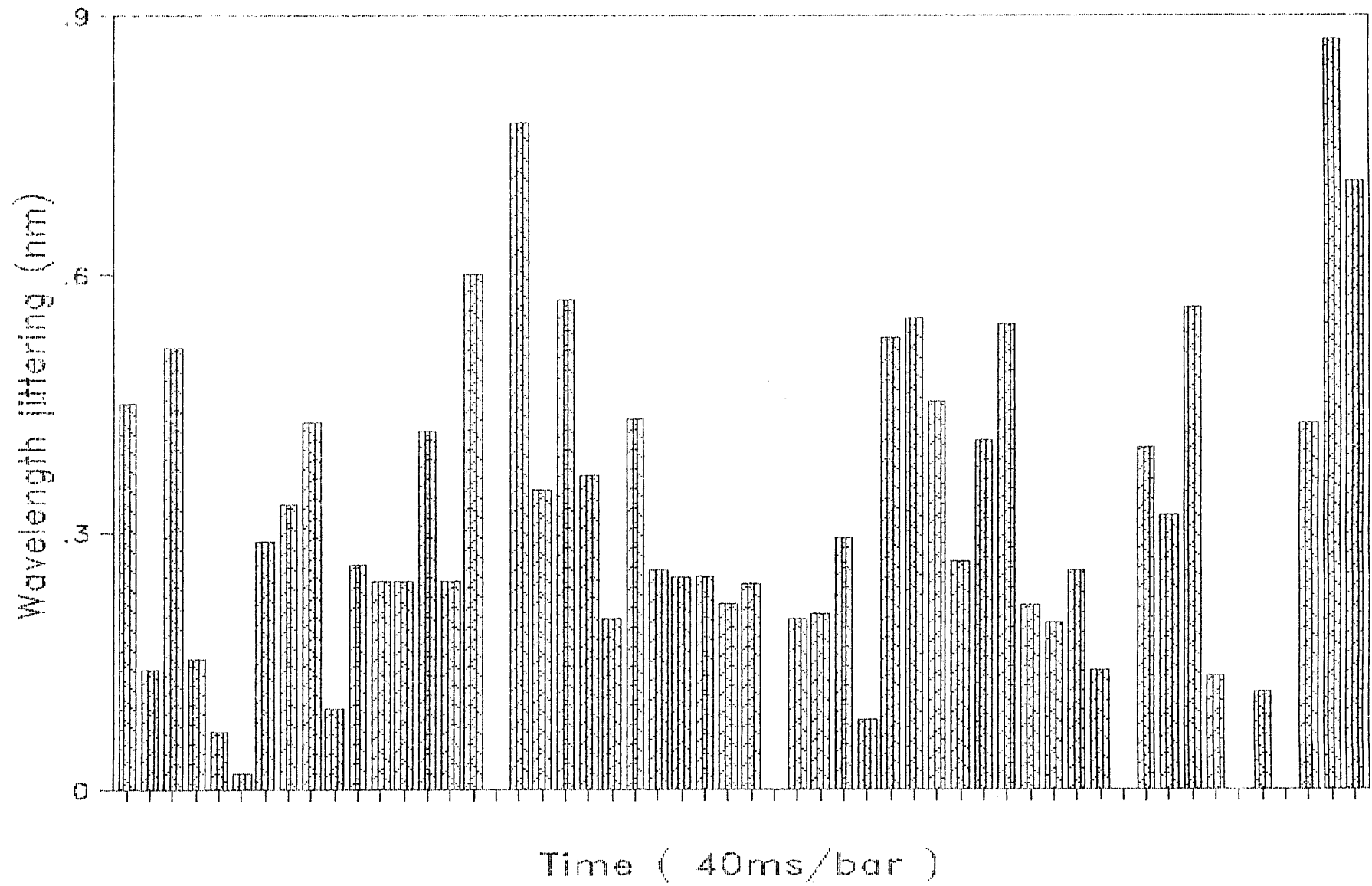


Fig. 4.10 Stable seed oscillator pulses wavelength jittering.

4.4 Stable Seed oscillator performance

In this chapter we have described how we have optimised and characterised the stability and performance of the seed oscillator. We have used a linear cavity with a prism pair for the intracavity dispersion compensation. The active jet fold section contains Coumarin 102 in benzyl alcohol and ethylene glycol as the gain medium, the two focusing mirrors each of 5 cm radius of curvature are aligned in a "Z" fold to compensate the off-axis astigmatism. The passive jet section contains DOCI in absolute alcohol and ethylene glycol as the saturable absorber, the jet is aligned at the focal point of mirror M_5 and at the centre of the radius of mirror M_4 to enhance the mode-locking mechanism. A dielectric coated tuning wedge is used as the wavelength tuning component because it is more stable in wavelength tuning than either a tuning slit or a focused beam pinhole tuning. With proper saturable absorber concentration the laser can operate at optimal efficiency. Under the proper operating conditions and by introducing suitable glass length in one of the prisms, we can compensate the intracavity group velocity dispersion, operate the laser in the stable regime and generate the shortest pulse duration.

The laser is tuned to 497.2 nm with characterised wavelength jitter of less than 0.6 nm. The output energy is typically greater than 10 mW and the pulse duration less than 250 fs. Pulses from this seed oscillator source have been satisfactorily amplified through the dye amplifiers chain, frequency doubled and further amplified in the KrF amplifier to generate high intensity ultrashort UV pulse.

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Chapter 5

Dye Amplifier Chain

In chapter 4 we described how stable seed oscillator pulses have been obtained from a passively mode-locked C102/DOCI dye laser with 10 mW output power at 140 MHz repetition rates and with 200 fs pulse durations. This source is tunable from 492 nm to 503 nm. The output energy per pulse is typically 70 pJ, corresponding to a peak power of 0.35 kW. It is necessary to amplify the seed pulse to much higher output energies, since for some particular applications powers in the MW and GW range are often required. The dye amplifier is capable of fulfilling these requirements because it is a high gain material. Typically more than 6 orders of magnitude gain can be achieved in the dye amplifier, boosting the output energy of the system up hundreds of μJ .

The amplifier dye usually flows in a cuvette or a Bethune cell^[5.1]. The dye amplifier can be pumped either longitudinally by a frequency doubled YAG laser^[5.2], or transversely by an excimer laser^[5.3]. Since the dye is a high gain material, a large amount of amplified spontaneous emission (ASE) will be generated together with the amplified seed pulse if the amplifier chain is not carefully configured to avoid it. ASE can seriously limit the gain of the seed pulses, and produce a pre-pulse as well as a pedestal on the amplified pulse. The dye amplifier chain is designed to ensure optimum amplification of the seed signal pulse with maximum energy extraction and

minimum ASE. Our approach is to employ multiple stages of dye amplifiers with spatial filters and/or saturable absorbers between the stages to eliminate ASE, and still obtain optimum amplification and maximum energy extraction.

5.1 Configuration of the dye amplifier chain

In a single stage dye amplifier it is difficult to control the ASE and it is difficult to have uniform transverse pumping because the interaction volume of the dye is necessarily large. With a multiple stage dye amplifier chain it is easy to eliminate ASE between stages and by optimising the dye volume in each stage and the relative pump beam ratio we can optimise the interaction volume to obtain best amplification and maximum energy extraction with minimum ASE.

Fig. 5.1 shows the configuration of the dye amplifier chain. The input seed beam is amplified by a 3 stage 4 pass dye amplifier transversely pumped by a XeCl excimer laser. The input seed beam is first focused through a large F number focusing lens L_1 . The first and second amplifier stages A_1 and A_2 are located symmetrically either side of the focal point of L_1 so that the seed beam fills the pumped volume and has the same diameter as it passes through the dye cells. A pinhole is positioned at the focal position of L_1 to eliminate the ASE from the first amplifier stage. After the second amplifier stage, A_2 , there is another spatial filter to eliminate the ASE from A_2 and a beam expander to expand the seed beam to a larger size for the final amplification and maximum energy extraction. The seed beam passes through a polarising cube and is amplified again in the third amplifier stage A_3 . The third

amplifier is in a double pass configuration via a quarter wave plate and a total reflector. The amplified output is finally coupled out from the amplifier chain through the polarising cube.

All three amplifier stages are Lambda Physik FL 443 dye circulators, consisting of power supply, dye pump, reservoir, filter and dye cell. The dye cell has a specially designed dye cuvette with rounded bottom that allows fast turbulent-free flow and so enables the system to be used at high repetition rates and high output powers. The dye cells in the A_1 and A_2 stages are of the same size, 1 mm in aperture and 2 cm in length. The dye cells are all mounted with maximum degree of freedom for alignment, and are deliberately tilted to the axis of the amplifier chain as shown in Fig. 5.2 in order to eliminate the possibility of laser oscillation between the optical surfaces. The dye cell in the A_3 stage is 2.5 mm in thickness and 4 cm in length. It is also tilted at an angle to suppress the oscillation and to restore the beam path height.

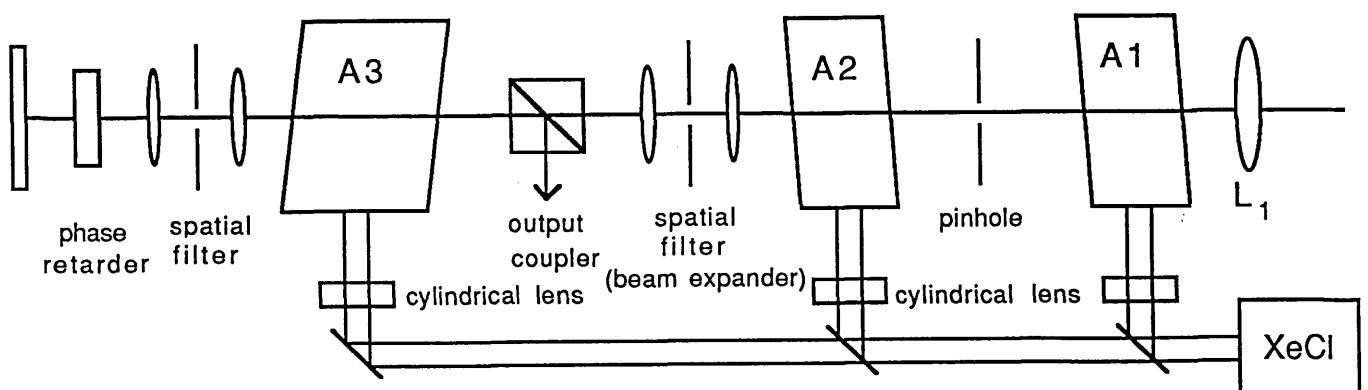


Fig. 5.1 Configuration of the dye amplifier chain.

All three amplifier stages are pumped by the same XeCl excimer laser (Lambda Physik, EMG 101 MSC). The pump beam is distributed to the first and second stage by two beam splitters and to the third stage by a total reflector. The distribution of the pump beam ratio in conjunction with the stage volume have significant effects on the amplification factors and the ASE ratio obtained. The pump beam is focused to the dye cells by cylindrical lenses.

5.2 Design consideration of the dye amplifier chain

The dye used in the amplifier stages is a high gain medium, which, although capable of providing huge amplification can also generate significant amounts of amplified spontaneous emission. Best advantage can be taken if a multi-stage amplifier is used and ASE is eliminated between each amplifier stage. In our set up, we use a 3 stage 4 pass amplifier. The basic idea is first to discriminate between the seed signal and the ASE signal in the first stage, and then to obtain best amplification in the following stage(s), with maximum energy extraction in the last stage.

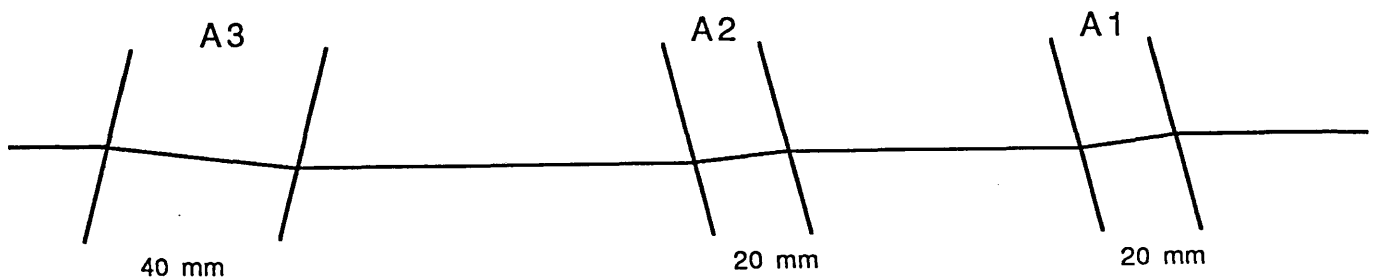


Fig. 5.2 The beam path of seed laser source through the amplifier chain.

It is extremely important that the amplified pulses after A_1 and the first pinhole should have good signal to ASE ratio, as shown in Fig. 5.3(a). If the amplified pulses are as shown in Fig. 5.3(b) and 5.3(c), then in the second and the third stages the ASE will only increase further, and will significantly suppress the gain in the signal pulse.

The second and third stages are the most important amplification stages. Maximum amplification should be obtained here. By proper optimisation, the seed pulses receive linear gain. When we double pass the seed beam through the last stage, we expect to operate the amplifier stage near the saturation edge since this will give maximum energy extraction and the output is less sensitive to the input intensity fluctuations.

The final design consideration is that it is essential to have synchronisation between the seed pulse relative to the peak of the pump beam to ensure best amplification. This is because the seed beam consists of an ultrashort pulse train of 7 ns separation (140 MHz) and the pulsewidth of the typical XeCl pump pulse is 30 ns.

5.2.1 Three stage four pass dye amplifier

The objective of the first stage of the dye amplifier is to discriminate the seed pulse signal and the ASE so that the signal to ASE ratio is maximised. The size of the A_1 stage is 1 mm in thickness and 2 cm in length. The small thickness and the shorter length ensure effective and homogeneous pumping.

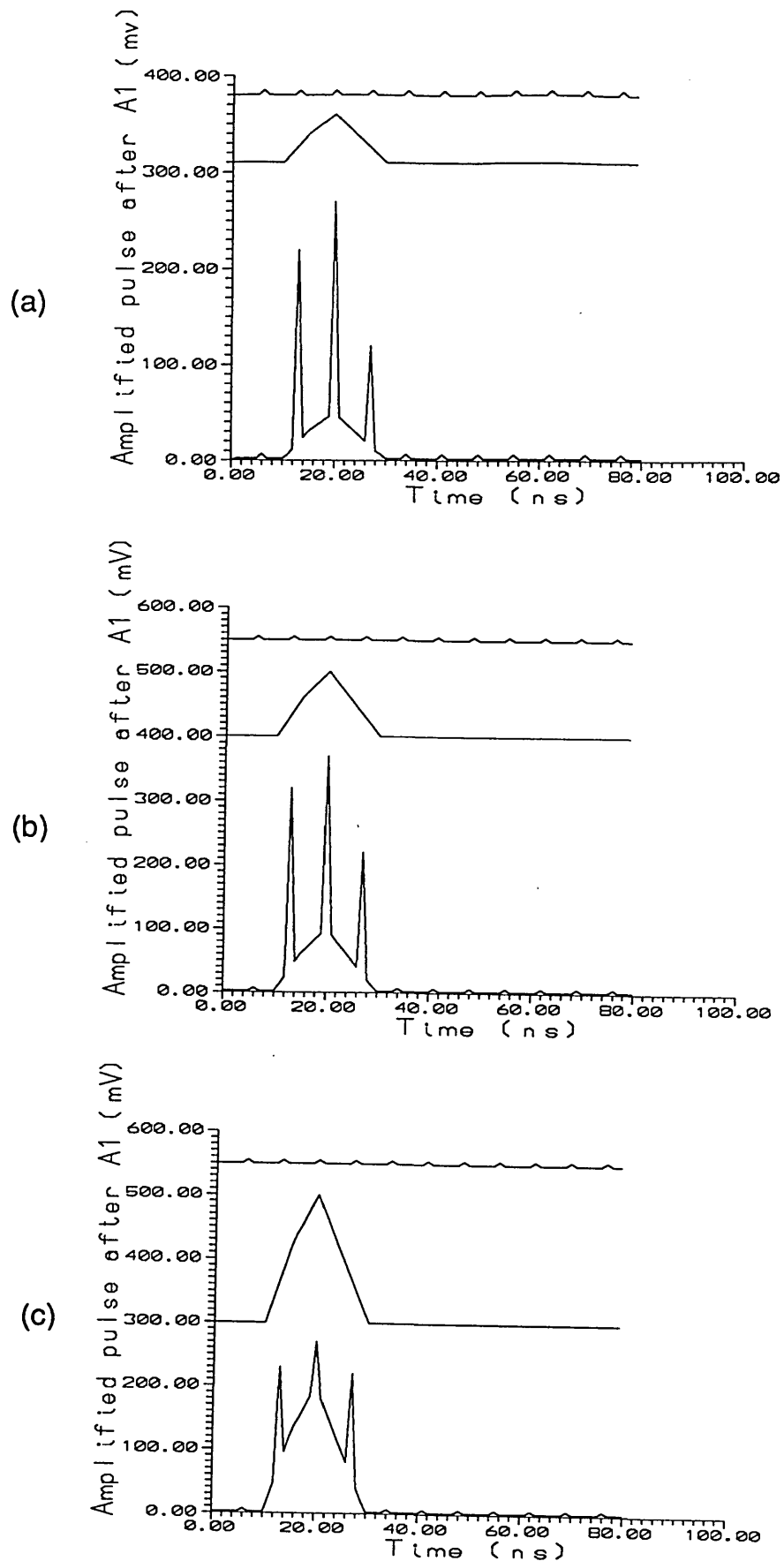


Fig. 5.3 Amplified seed pulses after A1 stage. (a) shows optimum amplification; (b) and (c) show the amplified pulses with increasing ASE levels. Upper trace is the seed pulses, central trace is the ASE pulse shape, lower trace is the amplified pulses with ASE pedestal.

By proper alignment of the overlap volume of the pump beam and seed beam volume, we have obtained rather satisfactory amplified signal to ASE ratios. The amplification gain in this stage is typically between 50 and 100.

The second stage of the amplifier, A_2 , is of the same size as A_1 . Since the amplified pulses from the A_1 are enhanced by nearly two orders of magnitude, the pulse will be amplified more significantly and typically overall gains of 10^4 to 10^5 have been obtained.

In the third stage, the thickness of the dye cuvette is 2.5 mm and the length is 4 cm. The pumping of this stage is much harder than the previous two stages, and the seed beam is expanded to a larger size for the best amplification and maximum energy extraction.

All three amplifier stages are aligned at angles tilted to the optics axis in order to eliminate oscillations. The seed beam path is shown in Fig. 5.2.

To take best advantage of the third amplifier to obtain amplification, maximum energy extraction and ASE elimination, we operate the third stage A_3 in a double pass configuration. By optimising the proper delay of the second pass within the high gain regime we can obtain more amplification and maximum energy extraction. An extra spatial filter could also be aligned in the double pass configuration to eliminate ASE more efficiently.

Corresponding to the different volume sizes of the seed beam and the different objectives of each stage, it is necessary to distribute the pump power in a suitable

ratio^[5.4]. In this system we use a 10 percent beam splitter to couple the first pump beam into A_1 , another 10 percent beam splitter to couple the second pump beam into A_2 , and a total reflector to direct all of the rest of the pump energy, 120 mJ, into the third stage.

ASE will inevitably accompany the amplification in the seed beam path. The purpose of the dye amplifier chain is to obtain the best amplification while keeping the ASE at a low level. This could be achieved by using spatial filtering and a saturable absorber such as Schott colour filter QC 530. A spatial filter is the most inexpensive, easiest to adjust and most effective method for controlling the ASE level.

The dye used in the amplifier chain is Coumarin 307. This is an efficient dye for amplifying in the blue-green. The peak absorption wavelength of Coumarin 307 is 395 nm, and the maximum fluorescence is at 488 nm. By optimising the dye concentrations in each stage in accordance with the seed beam size and the pump beam intensity, we can have the best operating conditions for the dye amplifier.

5.2.2 Pumping of the dye amplifier chain

The multistage dye amplifiers are transversely pumped by the XeCl excimer laser. The typical output waveform from the pump source is shown in Fig. 5.4. The thyatron in the XeCl laser was specially purchased because it has a maximum jitter of less than 2 ns as shown in fig. 5.4(b) for 20 shots. The output power of the pump source is typically 150 mJ and it can operate both in single shot manual mode and

up to 100 Hz repetition rates.

The objective of each dye stage is different and the seed beam volume as well as dye concentration are also different. It is necessary to properly distribute the pump power into each stage to meet the different requirements. The pump power distribution ratio should be optimised^[5.4]. In our system 10 percent beam splitters are used in both the first and second stages, and a total reflector is used at the last stage to couple all the remaining pump power into A_3 .

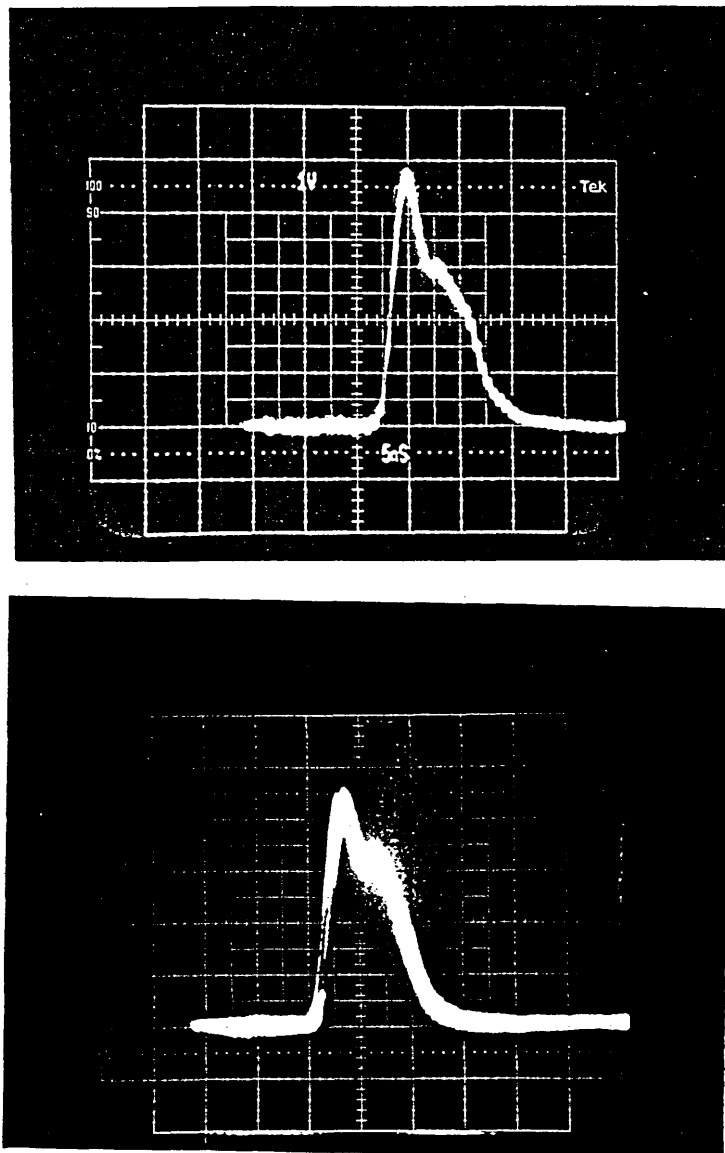


Fig. 5.4 Typical pulseshape of XeCl pump laser source. The lower photo shows the pump source jittered less than 2 ns (20 shots)

The pumping beams are focused into the dye cell via cylindrical lenses. The focal line position and the tilting angle of the cell have drastic effects on the amplification gain and the ASE generated in the seed beam path. Fig. 5.2 shows the beam path of the seed pulses. The pump beam should excite the seed beam volume as much as possible. If the pump beam is aligned in a direction exactly parallel and overlapping the seed beam path, the excited volume and interaction volume will be maximised. However, if the pump beam is parallel and on line with the seed beam path, the spontaneous fluorescence will also accumulate to a maximum level in the beam path direction despite the spatial filtering efforts along the amplifying axis. This causes a large amount of ASE that not only will overwhelm the amplification of the seed pulses in this stage but may deplete the gain in the following stage(s). Fig. 5.5(a) shows beam paths for the case when the pump beam is exactly parallel and on line with the seed beam path. This is not the situation we want. We should tilt the pump beam at an angle to the seed beam path. Although any tilting of the pump beam will reduce the interaction volume in the seed beam direction, this will also substantially reduce ASE and so enhance the amplification of the seed pulses. As the pump beam is tilted further, the interaction volume will change as shown in Fig. 5.5.

The pump beam should be optimised according to the objective of each stage. The cylindrical lens should also have maximum degree of freedom of alignment. The cylindrical lens is aligned as shown in Fig. 5.6(b) for minimum optical aberrations. For the optimum pumping of the seed beam volume, the focal line of the pump beam should be in the central region of the seed beam path as shown in Fig. 5.7(a). The

reflections of the pump beam from the rear surface of the dye cell can also help to keep the volume excited uniformly.

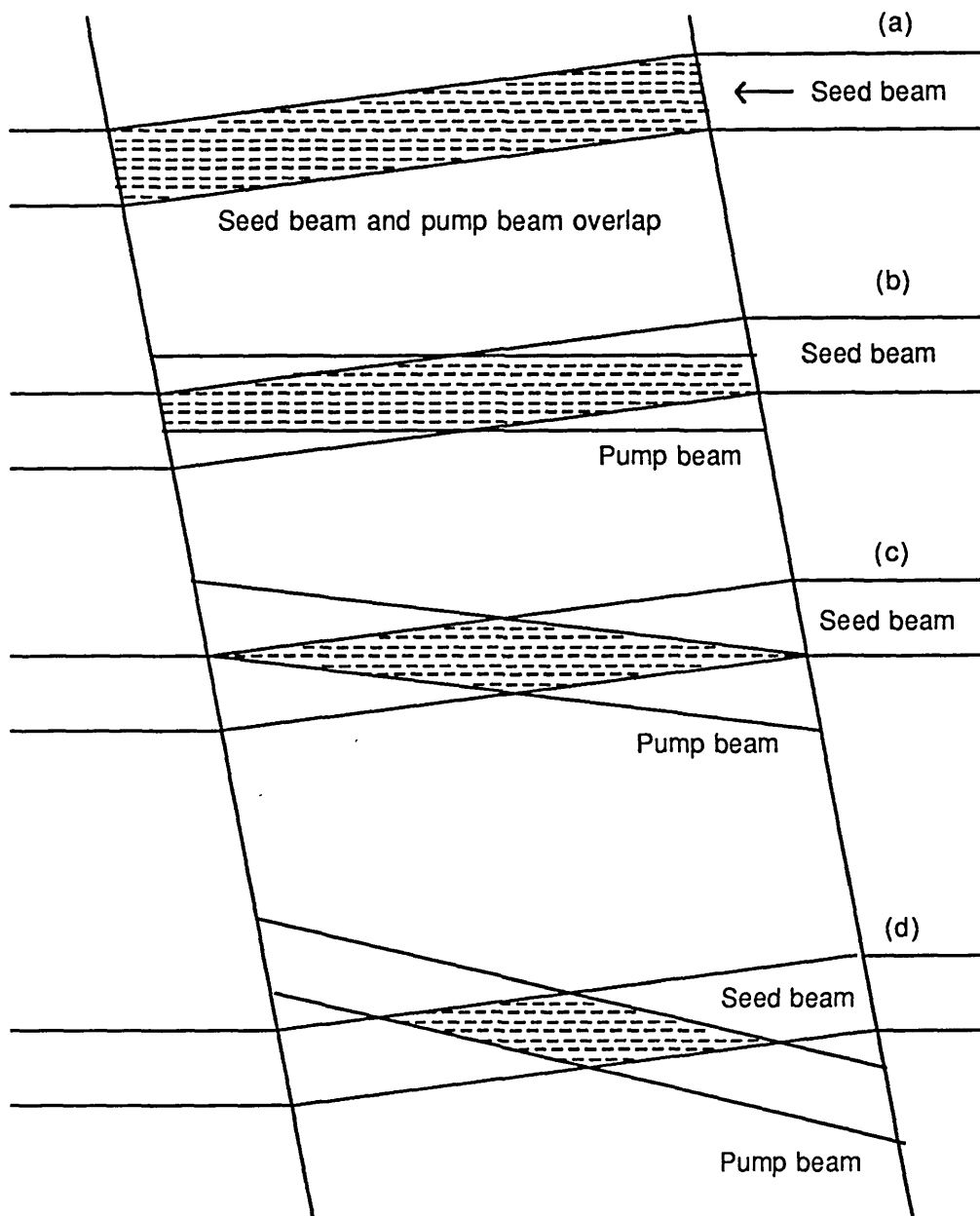


Fig. 5.5 Interaction volume of the pump beam and the seed beam. (a) shows the completely overlapping of the pump beam and the seed beam. (b) (c) (d) show the pump beam are tilted at different angles.

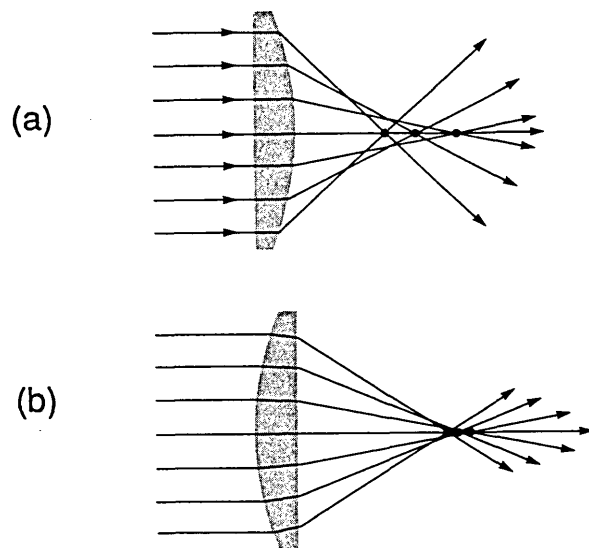


Fig. 5.6 The focusing of the cylindrical lens, (b) provides minimum optical aberration.

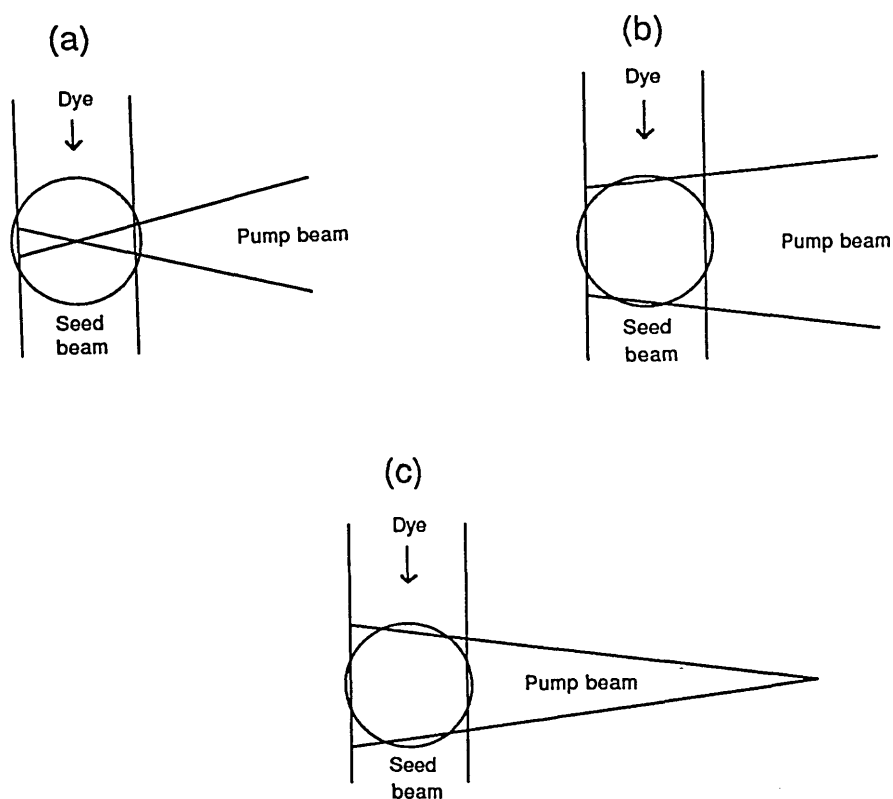


Fig. 5.7 The focusing of the pump beam into the dye cuvette. (a) shows the pump beam is focused to the central of the seed beam path and dye cuvette. (b) and (c) show the pump beam are focused beyond and in front of the dye cuvette.

5.3 Optimisations of the dye amplifier Chain

The objective of the first stage of the dye amplifier chain is to pre-amplify and minimise ASE. The objective in the second and third stages is to obtain best amplification, and finally in the last stage to obtain maximum energy extraction.

5.3.1 The amplification of the dye amplifier stages

The output of the amplified pulses after the first amplifier stage A_1 is shown in Fig. 5.8. The central trace shows the ASE shape only with input seed beam blocked. The ASE shape could be varied significantly by tilting the pump beam angle relative to the seed beam path and thus changing the interaction volume. By adjusting the relative timing of the pump and seed pulses we can synchronise the seed pulse to give best amplification. Higher amplified signal comes with higher ASE output, but if too much amplified spontaneous emission, the signal gain will be suppressed. Even with modest levels of ASE in the first stage output, and even if the following stages are aligned to minimise ASE, the ASE from the first stage will inevitably be amplified in the rest of the stages and limit the amplification of the seed signal. It is vital to optimise the output after the first amplifier stage to give minimum ASE and maximum amplification of the seed signal. Fig. 5.8 shows the optimized amplified signal. The amplification gain could be obtained in the range of 50 to 100.

When the amplified pulses come to the second stage a higher amplification factor is expected. Since the seed signal has already been enhanced to a higher level

it will accordingly be enhanced to a much higher level in the second stage, provided the gain in the second stage is not saturated. The stage size of A_2 is almost the same as A_1 , and with the same pump beam ratio. By proper alignment and optimisation, we can have amplified seed pulse as shown in Fig. 5.9. The amplification gain is in the range of 10^4 to 10^5 .

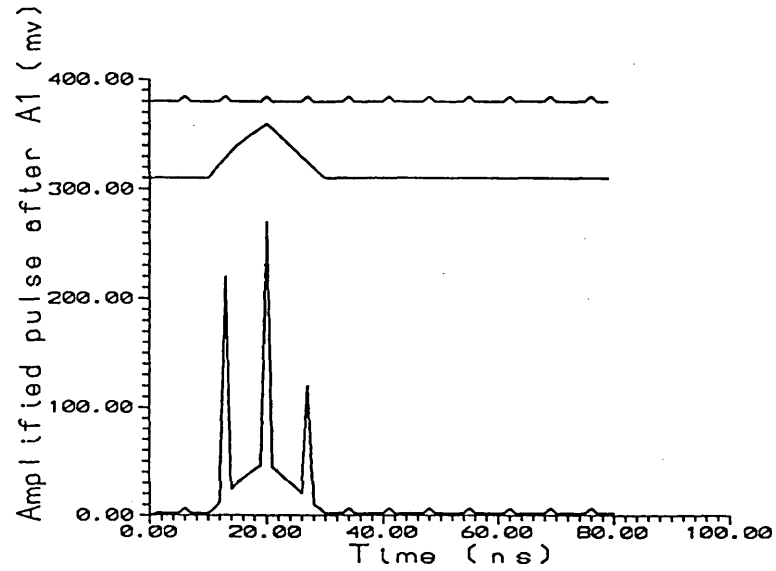


Fig. 5.8 Optimal amplified seed pulses after A1 amplifier stage.

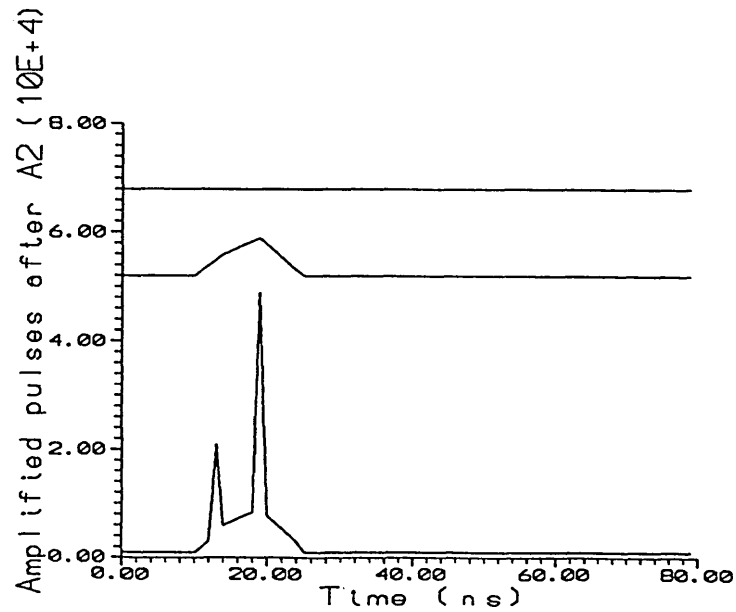


Fig. 5.9 Optimal amplified seed pulses after A2 amplifier stage.

The third stage is the last stage in our amplifier chain. We expect maximum energy extraction in this stage. From consideration of amplifier saturation and amplified signal energy extraction, it is advisable to expand the seed beam cross section. We have used the spatial filter after the A_2 stage and the expanding lens to expand the seed beam to 2.5 mm in diameter. In expanding the seed beam cross section we should take into consideration the dye amplifier size and the pump beam uniformity. Since the seed beam volume is increased, and we expect better energy extraction, we need all the remaining pump power. The pulse duration of the XeCl pump beam is 30 ns, and so we can also align a double passage in this last stage within the timescale of the pump. By optimising the delay time of the second pass, and installing a spatial filter after the first pass and a wave-plate for the polarised output coupling, the complete dye amplifier chain is as shown in Fig. 5.1. The overall amplification gain in the amplifier chain is 10^6 to 10^7 , and the output energy is in the range of 20 μJ to 100 μJ .

Overall the dye amplifier chain is operated near the edge of saturation to obtain maximum energy extraction and to minimise the output fluctuations. Fig. 5.10 shows the slope efficiency of the dye amplifier chain.

5.3.2 Amplified spontaneous emission (ASE) of the dye amplifier chain

To amplify the seed beam the amplifier medium must be pumped to an excited state and the excited volume overlapped by the seed beam path. The interaction volume provides gain amplification and also ASE signal in the seed beam direction.

ASE can not completely be eliminated. Fortunately, since the divergence angle of the ASE is much bigger then the seed beam, we can employ the spatial filters and the saturable absorber to control its ratio to the amplified signal.

The spatial filter could very effectively clean the amplified seed pulse, transmitting 99.3 percent of the signal energy^[5.5] at each stage if optimised. We have also used the Schott ~~filter~~ glass filter OG 530^[5.6] to control the ASE levels.

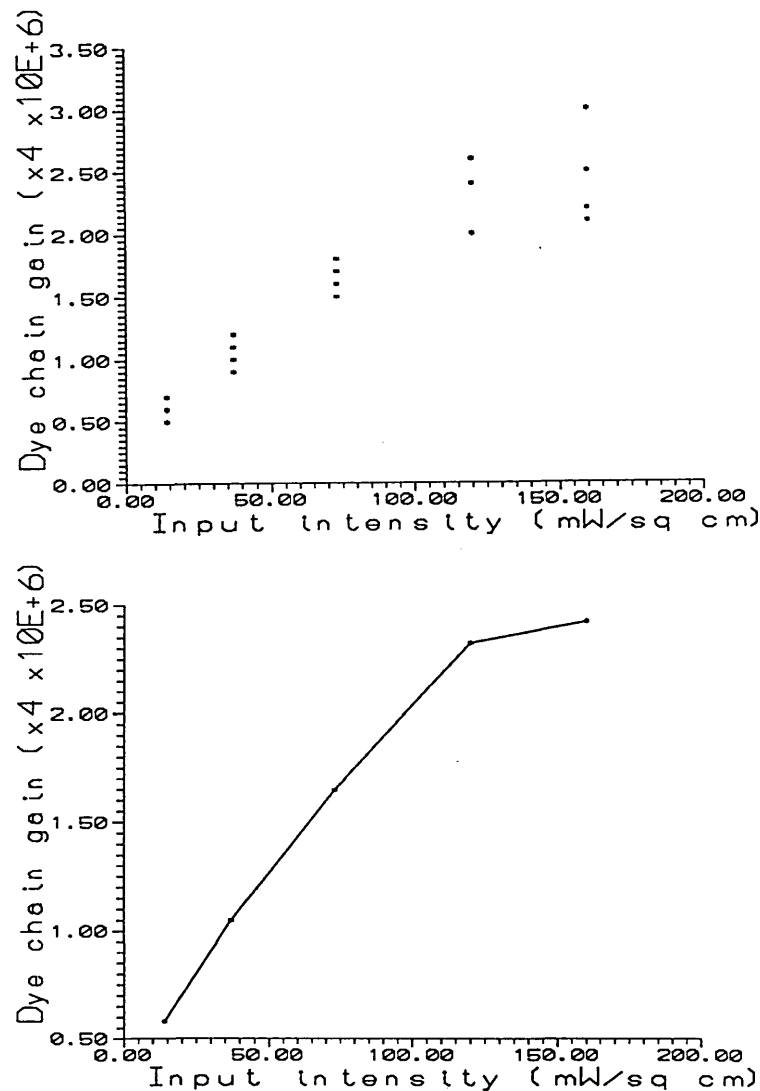


Fig. 5.10 Slope efficiency of the dye amplifier chain, lower figure shows the averaged data.

The typical ASE pulse shape after each amplifier is shown in Fig. 5.11. We can see that after passing through the three amplifier stages, the peak region of the ASE is enhanced more, and the rising edge as well as the falling edge become steeper.

The amount and the pulse shape of the ASE depends sensitively on the interaction volume of the seed beam and the pump beam as well as on the shape of the interaction volume. Different shapes of the same interaction volume can result in a faster rising slope and a slower trailing slope, or vice versa, or an equal lateral shape. The maximum width of the ASE shape is 10 ns after the third stage, as shown in Figure 5.1. Since the repetition rates of the seed pulse is 140 MHz, the separation between each seed pulse is approximately 7 ns. If we properly synchronise the timing, we can have the best amplified single pulse. The ASE shape could also be changed to be trapezoidal. That gives a little more ASE and decreases the amplification accordingly but is least sensitive to the system jittering. The best ASE shape is triangular with equal rising edge and falling edge, since the first amplified pulse will always obtain the best amplification and the second amplified pulse will not be amplified as much. This ensures that in the final stage only single pulse will be amplified significantly.

5.3.3 Synchronisation of the seed pulse train and the dye amplifier chain

The pulse duration of the XeCl pump beam is typically 30 to 40 ns. The ASE pulse shape after the first amplifier stage is 30 ns, after the second amplifier stage is 15 ns and after the double pass of the third amplifier stage is 10 ns. The seed beam is a mode-locked pulse train of 140 MHz repetition rate and so the separation of each

pulse in the seed beam is 7 ns. The optimal ASE pulsed shape is triangular. It is necessary to have synchronisation between the seed pulse train and the XeCl pump pulse to have the best single amplified pulse with maximum energy extraction. If we did not have proper synchronisation double amplified pulses with various timings and amplitudes would occur randomly.

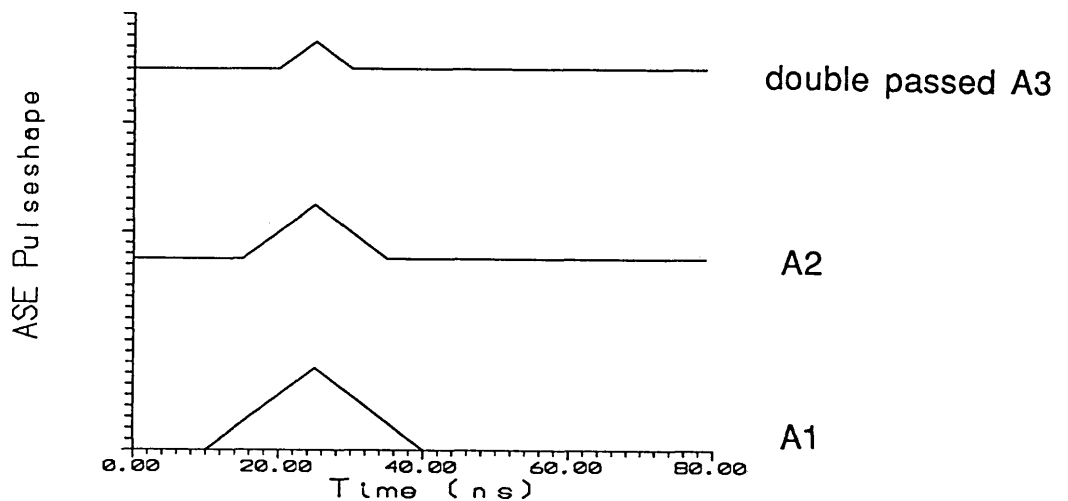


Fig. 5.11 ASE pulsed shape after A1, A2, and double passed A3 stage.
Each ASE pulsed shape is attenuated at different optical density.

Since the jitter of the CW passively mode-locked pulse train is of the order of a few tens of ps^[5,7], about two orders less than the maximum jitter of the thyatron of the XeCl pump source, we can quite comfortably neglect the seed pulse jitter and concentrate on the synchronisation of the seed beam relative to the peak of the ASE shape. The output of the seed pulse train on the fast response avalanche photodiode is amplified to trigger the TTL circuit and then trigger the SRS (Stanford Research System) delay signal generator to trigger the XeCl excimer pump source at a specific delay time. The SRS generator could also control the repetition rate of the pump source from manual single shot to several Hz and to a maximum of 100 Hz limited by the original design of the XeCl excimer laser.

5.3.4 Amplification in the dye amplifier chain

Following the objective design considerations we have achieved very satisfactory amplification in the dye amplifier chain. Table 5.1 shows the amplification and the output energy of the seed pulse after passing through the dye amplifier. Indeed, amplification of the order 10^6 to 10^7 , with output energy in the range of 20 μJ to 100 μJ has been provided. What is more, by optimisation of the interaction volume of the pump beam and seed beam, we can have single amplified pulse selection and obtain single amplified pulses without employing any pulse selection element such as a Pockels cell. It is most convenient and effective.

5.4 Characteristics of the dye amplifier chain

The dye amplifier chain can be optimised to amplify single femtosecond seed pulses with gain up to 10^6 and output energies of $50 \mu J$. The transition cross section σ of the dye amplifier is about $10^{-16} cm^2$, and the saturation energy E_s is $E_s = \frac{h\nu}{\sigma}$, equal to $2mJ/cm^2$. The output energy of the dye amplifier chain is in the range $50 \mu J$ to $100 \mu J$. Since the beam diameter is 2.5 mm, the energy flux is approximately equal to $2mJ/cm^2$. The dye amplifier is operated near the saturation regime for the best amplification and maximum energy extraction. Fig. 5.10 shows the slope efficiency of the dye amplifier chain. An additional advantage of operating the amplifier stage near the saturation region is that it is less sensitive to the input fluctuations.

Table 5.1 Gain and output energy after dye amplifier chain

	Gain	Saturation	Output Energy
A 1	50	No	3 nJ [*]
A 1 & A 2	2×10^4	No	700 nJ [*]
A 1, A 2, & A 3 double passed.	5×10^6	Yes	50 uJ

(* estimated value)

The gain depletion of the dye which implies high conversion efficiencies can also clearly be observed in the amplified pulseshape. Fig. 5.12 shows the typical gain depletion of the dye amplifier chain. We can see that the gain recovery time is of the order of 1 to 2 ns. In fig. 5.12 the lower trace is the amplified pulse with proper attenuation to the detector and the upper trace is the ASE only.

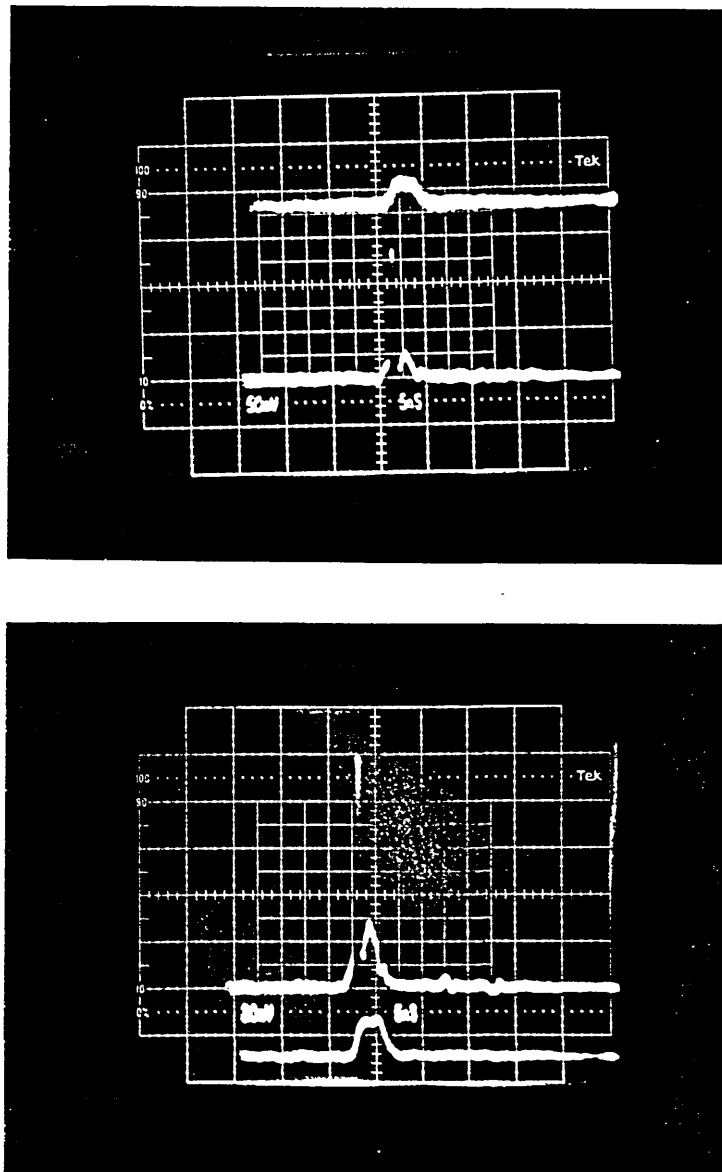


Fig. 5.12 Typical single amplified seed pulse after passing through the dye amplifier chain.

The ASE pulseshape is optimally aligned to have equal duration rising and trailing edge. Any synchronisation jittering will result in the seed pulse being amplified away from the peak of the amplification gain. We have deliberately deviated the synchronisation away from the peak and have found that before the double amplified pulses occur, a maximum deviation of ± 3 ns results in 50 percent decrease in the gain. Fig. 5.13 shows the results of the gain deviation of the dye amplifier chain.

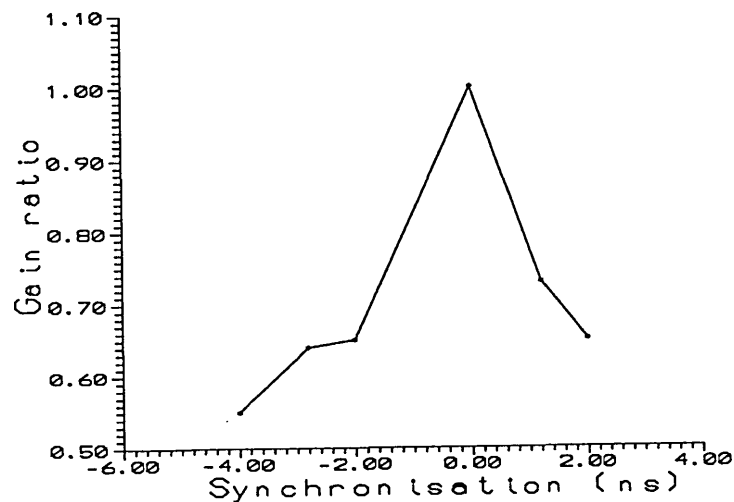


Fig. 5.13 Gain deviation of the dye amplifier chain with different synchronisation.

5.5 The tunable ultrashort pulse visible sub-system

The ultrashort pulse train from the passively mode-locked C102/DOCI laser of 10 mW output power at 140 MHz, 497.2 nm, and 200 fs pulse duration, corresponding to a peak power of 0.35 KW, have been amplified to output energies of 50 μJ , which correspond to peak powers of 160 MW, and peak power intensities of 4 GW/cm².

497.2 nm is the particular wavelength for the second harmonic generation of the KrF amplification. The seed source is tunable from 492 nm to 503 nm and the Coumarin 307 dye used in the dye amplifier has an even wider spectrum width.

The XeCl excimer pump source can operate at various repetition rates from manual single shot operation up to several Hz and a maximum 100 Hz.

Thus we have obtained an amplified tunable visible source subsystem of 50 μJ output energy, at peak powers of 160 MW and peak intensities of 3 GW/cm², focussable to 10¹³ W/cm² which is tunable from 492 nm to 503 nm.

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Chapter 6

Second Harmonic Generation and KrF Amplification

In chapter 5, we described how the ultrashort pulse train from the stable seed oscillator is amplified when passing through the dye amplifier chain. Single amplified pulses of 497.2 nm, 50 μJ output energy, and peak power density of 2 GW/cm² can be obtained. The amplified pulse is tunable from 488 nm to 503 nm and can operate at high repetition rates up to 100 Hz. Thus in chapter 5, we have described a useful high repetition rate ultrashort pulse visible source subsystem.

Applications such as high order harmonic generation, x-ray and plasma physics investigations require high peak intensities in the UV regime and in this chapter we describe how this is achieved. We can frequency double the 497.2 nm pulse to 248.6 nm, and amplify the pulse through a powerful KrF amplifier. The KrF source can lase and be used as an amplifier in 248 nm. Its operating wavelength, 248 nm, is shorter than that of XeCl excimer which is 308 nm. It has a higher saturation energy of 2 mJ/cm² compared to that of 0.85 mJ/cm² for the XeCl source^[6.1] and it can easily be scaled up to much larger discharge volumes. What is more, it has a broad gain bandwidth that can support the amplification of ultrashort pulses of a few tens of femtoseconds duration^[6.2]. All these factors make the KrF source the best candidate for achieving a high intensity, ultrashort pulse, UV laser system. The excimer source can operate at high repetition rates and this gives another advantage for various

experimental application.

6.1 Second harmonic generation

Our tunable visible source can stably operate at 497.2 nm, that is exactly double the operating wavelength of the KrF amplifier.

There are several second harmonic generation (SHG) crystals capable of generating harmonic pulses in the UV regime^[6.3]. Among these, the BBO (Beta Barium Borate $\beta - BaB_2O_4$) crystal is the most distinguished one.

Imagine a nonlinear medium driven by an optical field of frequency ω

$$E(t) = Re(E_\omega e^{-i\omega t}) = \frac{1}{2}[E_\omega e^{-i\omega t} + E_\omega^* e^{i\omega t}] \quad (6.1)$$

Take this to be a plane wave travelling in the Z-direction

$$E = \frac{1}{2}[E_\omega(Z)e^{-i(\omega t - k_\omega Z)} + E_\omega^*(Z)e^{+i(\omega t - k_\omega Z)}] \quad (6.2)$$

Where $k_\omega = \frac{n(\omega)\omega}{c}$ and $n(\omega)$ is the linear refractive index.

Write the polarisation to the second order as^[6.5]

$$\begin{aligned} P(Z, t) = & \frac{1}{2}[P_\omega^{(1)} e^{-i(\omega t - k_\omega Z)} + P_\omega^{(1)*} e^{+i(\omega t - k_\omega Z)}] \\ & + \frac{1}{2}[P_{2\omega}^{(2)} e^{-2i(\omega t - k_\omega Z)} + P_{2\omega}^{(2)*} e^{+2i(\omega t - k_\omega Z)}] \end{aligned} \quad (6.3)$$

where we have ignored the optical rectification term $\chi^{(2)}E_\omega^2$ and

$$P_\omega^{(1)} = \epsilon_0 \chi^{(1)}(\omega) E_\omega(Z) \quad (6.4)$$

$$P_{2\omega}^{(2)} = \epsilon_0 \chi^{(2)}(\omega) E_\omega^2(Z) = \bar{d} E_\omega^2(Z) \quad (6.5)$$

represent the linear refractive index term and the quadratic second harmonic generation (SHG) term respectively.

This polarisation acts as a source in Maxwell's wave equation

$$\nabla^2 E - \epsilon_0 \mu_0 \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2} \quad (6.6)$$

We write the SHG field as

$$E(SHG) = \frac{1}{2} [E_{2\omega}(Z) e^{-i(2\omega t - k_{2\omega} Z)} + c.c.] \quad (6.7)$$

Where $k_{2\omega} = \frac{n(2\omega)2\omega}{c}$ and $n(2\omega)$ is the linear refractive index of the medium for light of frequency 2ω .

If we assume $E_{2\omega}(Z)$ is slowly-varying with Z compared with $e^{ik_{2\omega} Z}$, we can neglect the second derivatives of $E_{2\omega}(Z)$ in equation (6.6) and for plane wave propagation i.e, $\nabla^2 E = \frac{\partial^2 E}{\partial Z^2}$, we can write the SHG growth equation as

$$\frac{dE}{dZ}_{2\omega} = i\omega\left(\frac{\mu_0}{\epsilon_{2\omega}}\right)^{\frac{1}{2}} \bar{d}E^2_{\omega}(Z)e^{i\Delta k Z} \quad (6.8)$$

where

$$\begin{aligned} k_{2\omega}^2 &= (2\omega)^2 \epsilon_{2\omega} \mu_0 = 4\omega^2 \epsilon_0 \mu_0 n^2(2\omega) \\ &= 4\omega^2 \epsilon_0 \mu_0 [1 + \chi^{(1)}(2\omega)] \end{aligned} \quad (6.9)$$

and the wave-vector, or photon momentum mismatch Δk is

$$\Delta k = 2k_{\omega} - 2k_{2\omega} = 2\omega(\epsilon_0 \mu_0)^{\frac{1}{2}} [n(\omega) - n(2\omega)] \quad (6.10)$$

In a normal dispersive medium $n(2\omega) \geq n(\omega)$. To solve the SHG growth equation we need to know $E_{\omega}(Z)$. We assume a low conversion holds, ie the depletion of $E_{\omega}(Z)$ from the initial value $E_{\omega}(0)$ is negligible, and $E_{\omega}(Z) \approx E_{\omega}(0)$. Then equation (6.8) is simply integrated to give

$$E_{2\omega}(Z) = i\omega\left(\frac{\mu_0}{\epsilon_{2\omega}}\right)^{\frac{1}{2}} \bar{d}E^2_{\omega}(0)Z e^{i\frac{1}{2}\Delta k Z} \left(\frac{\sin \frac{1}{2}\Delta k Z}{\frac{1}{2}\Delta k Z}\right) \quad (6.11)$$

If the nonlinear medium responsible for the SHG has a length L , the output electric field at 2ω at L is $E_{2\omega}(Z = L)$.

The time-averaged intensities at ω and 2ω are given by

$$I_{\omega} = \frac{1}{2} \left(\frac{\epsilon_{\omega}}{\mu_0} \right)^{\frac{1}{2}} |E_{\omega}|^2 \quad (6.12)$$

$$I_{2\omega} = \frac{1}{2} \left(\frac{\epsilon_{2\omega}}{\mu_0} \right)^{\frac{1}{2}} |E_{2\omega}|^2 \quad (6.13)$$

So the SHG intensity at the exit face is

$$I_{2\omega}(L) = 2 \left(\frac{\mu_0}{\epsilon_0} \right)^{\frac{3}{2}} \frac{\bar{d}^2 \omega^2}{n^2(\omega)n(2\omega)} I_{\omega}^2(0) L^2 \left(\frac{\sin \frac{1}{2} \Delta k L}{\frac{1}{2} \Delta k L} \right)^2 \quad (6.14)$$

That is, the SHG intensity is proportional to : $\bar{d}^2 = \epsilon_0(\chi^{(2)})^2$; the pump intensity $(I_{\omega})^2$; length $(L)^2$ of the SHG medium; and a rapidly-varying term in Δk .

The SHG intensity for fixed Δk at first grows quadratically with distance L , but maximises after a coherence length, l_{coh}

$$l_{coh} = \frac{\pi}{\Delta k} = \frac{c\pi}{2\omega} [n(\omega) - n(2\omega)]^{-1} \quad (6.15)$$

and thereafter the energy in the SHG wave is returned to the pump wave until the process starts all over again. Therefore it is of no value to use a medium of length $L \geq l_{coh}$ because the output will come from the last coherence length. The index mismatch responsible for the effect is due to the pump wave overtaking the harmonic wave or vice versa, and getting out of phase. The SHG is maximised if the medium is index matched with $n(2\omega) = n(\omega)$ and $\Delta k = 0$. If this is true then

$$\left(\frac{\sin \frac{1}{2} \Delta k Z}{\frac{1}{2} \Delta k Z}\right)^2 \rightarrow 1 \quad (6.16)$$

and the maximum power conversion is

$$\eta_{max SHG} = \frac{I_{2\omega}(L)}{I_{\omega}(0)} = 2\left(\frac{\mu_0}{\epsilon_0}\right)^{\frac{3}{2}} \frac{\bar{d}^2 \omega^2}{n^2(\omega)n(2\omega)} I_{\omega}(0) L^2 (1) \quad (6.17)$$

Generally the SHG efficiency is expressed as

$$\eta_{SHG} = \frac{I_{2\omega}(L)}{I_{\omega}(0)} = 2\left(\frac{\mu_0}{\epsilon_0}\right)^{\frac{3}{2}} \frac{\bar{d}^2 \omega^2}{n^2(\omega)n(2\omega)} I_{\omega}(0) L^2 \left(\frac{\sin \frac{1}{2} \Delta k L}{\frac{1}{2} \Delta k L}\right)^2 \quad (6.18)$$

we notice that the conversion efficiency from ω to 2ω is proportional to the input intensity $I_{\omega}(0)$, and $I_{\omega}(0) \equiv \frac{P_{\omega}(0)}{A} = \frac{1}{2} \sqrt{\frac{\epsilon}{\mu}} |E^{(\omega)}|^2$ of the fundamental beam. Also

$$\eta_{SHG} = \frac{P_{2\omega}(L)}{P_{\omega}(0)} = 2\left(\frac{\mu_0}{\epsilon_0}\right)^{\frac{3}{2}} \frac{\bar{d}^2 \omega^2}{n^2(\omega)n(2\omega)} \frac{P_{\omega}(0)}{A} L^2 \left(\frac{\sin \frac{1}{2} \Delta k L}{\frac{1}{2} \Delta k L}\right)^2 \quad (6.19)$$

The analysis of second harmonic generation leading to equations (6.18) and (6.19) is based on a plane wave model^[6,4]. In practice one uses a Gaussian beam which is focused so as to reach its minimum radius (waist) inside the SHG crystal. The incident Gaussian beam is characterised by the confocal parameter z_0 , which is the distance from the beam waist in which the beam area $\pi \omega^2$ is double that of the waist. $z_0 = \frac{\pi \omega_0^2 n}{\lambda}$ where ω_0 is the minimum beam radius (waist).

If $z_0 \gg L$, where L is the SHG crystal length, the beam area and hence the intensity of the incident wave is nearly independent of z within the SHG crystal. Since

$$|E^{(2\omega)}(r)|^2 = \frac{\mu}{\epsilon} \omega^2 \bar{d}^2 |E^{(\omega)}(r)|^4 L^2 \left(\frac{\sin \frac{1}{2} \Delta k L}{\frac{1}{2} \Delta k L} \right)^2 \quad (6.20)$$

where

$$E^{(\omega)}(r) = E_0 e^{-\left(\frac{r^2}{\omega_0^2}\right)} \quad (6.21)$$

as appropriate for a fundamental Gaussian beam, and using

$$P^{(\omega)} = \frac{1}{2} \sqrt{\frac{\epsilon}{\mu}} \int_{\text{crosssection}} |E^{(2\omega)}|^2 dx dy \quad (6.22)$$

and equation (6.21) and integrating equation (6.20), we have

$$\frac{P_{2\omega}(L)}{P_{\omega}(0)} = 2 \left(\frac{\mu_0}{\epsilon_0} \right)^{\frac{3}{2}} \frac{\bar{d}^2 \omega^2}{n^2(\omega) n(2\omega)} \left(\frac{P_{\omega}(0)}{\pi \omega_0^2} \right) L^2 \left(\frac{\sin \frac{1}{2} \Delta k L}{\frac{1}{2} \Delta k L} \right)^2 \quad (6.23)$$

The results in equation (6.23) is identical to that of equation (6.19), however it is derived for a Gaussian beam input with $z_0 \gg L$. From this result, at a fixed crystal length L and a given input power $P^{(\omega)}$ the SHG efficiency can be increased by decreasing the beam waist ω_0 . This is indeed true until z_0 becomes comparable to L . Further reduction of the beam waist ω_0 , and accordingly z_0 since $z_0 = \frac{\pi \omega_0^2 n}{\lambda}$, will lead to a situation in which the beam begins to spread appreciably within the crystal, thus leading to a reduced intensity and a reduced second harmonic generation.

It is thus reasonable to focus the beam until $L = 2z_0$. At this point $\omega_0^2 = \frac{\lambda L}{2\pi n}$,

which is referred to as confocal focusing and the optimal second harmonic generation efficiency is^[6.4]

$$\begin{aligned}\eta_{\text{confocal focusing}} &= \frac{P_{2\omega}(L)}{P_{\omega}(0)_{\text{confocal focusing}}} \\ &= \frac{2}{\pi c} \left(\frac{\mu_0}{\epsilon_0} \right)^{\frac{3}{2}} \frac{\bar{d}^2 \omega^2 L}{n^2} P_{\omega}(0) \left(\frac{\sin \frac{1}{2} \Delta k L}{\frac{1}{2} \Delta k L} \right)^2\end{aligned}\quad (6.24)$$

The main difference between the confocal focusing of the Gaussian beam and the plane wave results is that the conversion efficiency in this case increases as L instead of L^2 . This reflects the fact that a longer crystal entails the use of a larger beam spot size ω_0 so as to keep $z_0 \approx \frac{L}{2}$, which therefore reduces the intensity of the input fundamental beam.

In the practical high conversion efficiency case we can not still assume that the input intensity at ω is not affected by the SHG interaction. The exact solution is substantially saturated instead of the monotonically increasing solution given by the no depletion approximation^[6.4]. In the extreme case, as ~~that~~ all the input photons can be converted into half as many output photons at twice the frequency and no more.

Finally, we still need to include the real Δk ^[6.4]. Consider ~~for~~ a typical BBO crystal : $n(497.2 \text{ nm}) = 1.6777$, but $n(248.6 \text{ nm}) = 1.7793$; so $\Delta k = \frac{2\omega}{c}(n(\omega) - n(2\omega)) = 2.4 \times 10^6 \text{ m}^{-1}$, and $\Delta k L = 3.6 \times 10^3$, for $L = 1.5 \text{ mm}$. The ~~sin~~² factor in equations (6.18) and (6.19) gives $\left(\frac{\sin 3600}{3600} \right)^2 \simeq 5 \times 10^{-9}$. And $\eta_{SHG} < 10^{-10}$ instead. Clearly we have to find a way of making $\Delta k \approx 0$, ie by phase-matching.

The difference in phase velocities between ordinary and extraordinary waves in uniaxial media can be used to phase match pump and harmonic waves. The dependence of the index of refraction of the extraordinary wave in a uniaxial crystal on the angle θ between the propagation direction and the crystal optics (Z) axis is given as^[6.4]

$$\frac{1}{n_e^2(\theta)} = \frac{\cos^2 \theta}{n_o^2} + \frac{\sin^2 \theta}{n_e^2} \quad (6.25)$$

If $n_e^{2\omega} \leq n_o^\omega$, there exists an angle θ_m at which $n_e^{2\omega}(\theta_m) = n_o^\omega$; so if the fundamental beam at ω is launched along θ_m as an ordinary ray, the second harmonic beam will be generated along the same direction as an extraordinary ray. ~~The situation is illustrated in Fig. 6.1.~~ The angle θ_m is determined by the intersection between the sphere corresponding to the index surface of the ordinary beam at ω with the index surface of the extraordinary ray which gives $n_e^{2\omega}(\theta)$. The angle θ_m for negative uniaxial crystals, that is crystals in which $n_e^\omega \leq n_o^\omega$ is that satisfying $n_e^{2\omega}(\theta_m) = n_o^\omega$, which from equation (6.25) is

$$\frac{\cos^2 \theta_m}{(n_o^{2\omega})^2} + \frac{\sin^2 \theta_m}{(n_e^{2\omega})^2} = \frac{1}{(n_o^\omega)^2} \quad (6.26)$$

solving for θ_m

$$\sin^2 \theta_m = \frac{(n_o^\omega)^{-2} - (n_o^{2\omega})^{-2}}{(n_e^{2\omega})^{-2} - (n_o^{2\omega})^{-2}} \quad (6.27)$$

6.2 BBO second harmonic generation

Beta barium borate (β -BBO) is a new nonlinear optical material discovered in 1984^[6.6]. It is an interesting material characterised by a wide range of transparency in both the ultraviolet and the near infrared. Its large birefringence allows phase-matching for many harmonic generation processes, and in particular it has been used to generate the fifth harmonic in Nd:YAG^[6.3]. It has a substantial nonlinear coupling. These properties indicate that the barium borate is potentially useful for many applications, especially in the ultraviolet region below 250 nm. In addition, BBO has a high damage threshold, is physically robust, and has excellent average power capabilities.

The refractive indices of barium borate can be obtain from the Sellmeier form of^[6.7]

$$n^2 = A + B/(\lambda^2 + C) + D\lambda^2 \quad (6.28)$$

and Table 6.1 gives the Sellmeier constants obtained^{[6.6][6.7][6.8]}. So the Sellmeier equations of the BBO are

$$n_e^2 = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2$$
$$n_o^2 = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2 \quad (6.29)$$

Table 6.2 lists the refractive indices at the wavelength of our seed oscillator, 497.2 nm and at the KrF wavelength 248.6 nm.

In the general three-wave mixing experiment, waves at frequencies ω_1 and ω_2 are mixed to generate a wave at $\omega_3 = \omega_1 + \omega_2$. The simple plane wave theory, neglecting depletion, predicts that the output intensity is

$$I \propto I_0 \sin^2(\delta) \quad (6.30)$$

where δ is the phase mismatch, $\delta = \frac{1}{2}\Delta k L$. L is the crystal thickness and Δk is the wave-vector mismatch,

$$\Delta k = k_3 - k_2 - k_1 = \frac{1}{c}(n_3\omega_3 - n_2\omega_2 - n_1\omega_1) \quad (6.31)$$

The refractive indices depend on the polarisation of the waves, and in general there are three different polarisation configurations which can lead to the vanishing phase mismatch. In optical materials there are two propagating modes of the electromagnetic field at each frequency. In a birefringent medium they have different velocities and orthogonal polarisations. One polarisation is fast, the lower refractive index; and the other is slow, the higher refractive index. In biaxial crystals, both velocities depend on the direction of propagation of the wave through the crystal. They are both considered extraordinary waves. However for uniaxial crystals such as barium borate only one velocity varies with propagation direction. Since for the barium borate which is negatively birefringent, $n_e^\omega = 1.5571 \leq n_o^\omega = 1.6777$, the fast

velocity depends on the propagation direction and is the extraordinary wave.

With the convention that $\omega_1 \leq \omega_2 \leq \omega_3$, the three types of phase matching are summarized in Table 6.3. For frequency doubling type 3 and type 2 are the same process, whereas this is not the case for sum frequency generation. In this system we used type 1 phase matching.

The phase matching angle θ_m for a particular mixing process at a reference temperature T_0 is given by the following implicit equation^[6.7]

$$\Delta k(\theta_m, T_0) = 0 \quad (6.32)$$

For angles near θ_m and temperatures near T_0 the wave-vector mismatch Δk can be expanded as a power series in θ and T as

$$\Delta k = \beta_\theta(\theta - \theta_m) + \beta_T(T - T_m) \quad (6.33)$$

where $\beta_\theta = \frac{\partial \Delta k}{\partial \theta}$, and $\beta_T = \frac{\partial \Delta k}{\partial T}$. The angular sensitivity β_θ and the temperature sensitivity β_T can be derived directly from equation (6.31) by calculating the refractive index data. Since the barium borate has lower temperature sensitivity^[6.7], we shall concentrate only on the angular phase matching.

Using equation (6.27) and the refractive index data of Table 6.3 we can calculate the phase matching angle $\theta_m = 52.8^\circ$.

Table 6.1 Sellmeier parameters for BBO crystal

	A	B (μm^2)	C (μm^2)	D (μm^{-2})
n_e	2.3753	0.01224	- 0.01667	- 0.01516
n_o	2.7359	0.01878	- 0.01882	- 0.01354

Table 6.2 Refractive indices of BBO crystal at KrF harmonics

$\lambda(\mu m)$	n_o	n_e
497.2	1.6777	1.5571
248.6	1.7793	1.6265

Table 6.3 Phase-matching type in BBO, e = extraordinary wave (faster)
o = ordinary wave (slower)

Type	ω_1	ω_2	ω_3
1	o	o	e
2	e	o	e
3	o	e	e

The second harmonic generation conversion efficiency can be estimated from equations (6.19) and (6.24). This depends on the intensity of the input waves and also on the phase mismatch between them. The crystal length is limited by the angular sensitivity of the process and the beam divergence. Therefore there is a minimum peak intensity required to achieve efficient conversion. Note that for efficient harmonic generation the peak power of the fundamental laser beam must exceed the threshold by a significant margin between 1 and 10. That is, the minimum threshold power intensity for efficient type 1 second harmonic generation at $0.5 \mu m$ is about $100 MW/cm^2$ ^[6,7]. Also the peak power required increases by Q^2 , where Q is the beam quality, approximately the ratio of the beam divergence to the divergence of a diffraction-limited laser beam.

The output energy of the seed pulse after the dye amplifier is typically of $50 \mu J$ at the pulse duration of 300 fs and 2.5 mm in diameter of the beam spot. That gives a peak power intensity of $800 MW/cm^2$. Using equation (6.19) for the plane wave approximation for the second harmonic generation of the BBO crystal, with

$$\mu_0 = 4\pi \times 10^{-7} H/m$$

$$\epsilon_0 = 8.854 \times 10^{-12} F/m$$

$$\lambda = 497.2 nm = 0.4972 \times 10^{-6} m$$

$$\omega = 2\pi\nu = 2\pi \frac{c}{\lambda} = 3.79 \times 10^{15} Hz$$

$$\bar{d} = 1.6 \times 10^{-23} mks$$

$$n_0^{(497.2nm)} = 1.6777$$

$$n_0^{(248.6nm)} = 1.7793$$

$$L = 1mm = 1 \times 10^{-3}m$$

$$I = 8 \times 10^8 W/cm^2 = 8 \times 10^{12} W/m^2$$

and

$$\eta_{SHG} = \frac{P_{2\omega}(L)}{P_{\omega}(0)} = 2\left(\frac{\mu_0}{\epsilon_0}\right)^{\frac{3}{2}} \frac{\bar{d}^2 \omega^2}{n^2(\omega)n(2\omega)} \frac{P_{\omega}(0)}{A} L^2 \left(\frac{\sin \frac{1}{2} \Delta k L}{\frac{1}{2} \Delta k L}\right)^2 \quad (6.19)$$

so

$$\begin{aligned} \eta_{SHG} &= 2 \times \left(\frac{4\pi \times 10^{-7}}{8.854 \times 10^{-12}}\right)^{\frac{3}{2}} \times \frac{(3.79 \times 10^{15})^2 \times (1.6 \times 10^{-23})^2}{(1.6777)^2 (1.7793)} \times 10^{-3} \times 8 \times 10^{12} \times 1 \\ &= 6.28 \times 10^{-1} = 0.628 \end{aligned}$$

The estimated SHG efficiency of 62 percent is somewhat optimistic since there are always angular and temperature phase mismatches, bandwidth limitation on the ultrashort pulses, input power intensity degradation, and optical Fresnel losses etc. We have measured the SHG efficiency after the dye amplifier and the BBO crystal to be 4 percent, and typically 2 to 3 percent efficiency could be obtained routinely. Schafer et al^[6.2] reported 8 percent conversion efficiency using a 0.2 mm thick, 52° cut BBO crystal in their system.

The second harmonic generation could also improve the S/N ratio of the amplified seed pulse to the dye amplifier ASE. This is because the peak intensity of the

amplified seed pulse is at least 3 orders of magnitude greater than the dye amplifier ASE.

The length of the crystal was carefully chosen, since it has to meet conflicting requirements. In order to match the phase-matched bandwidth of the crystal with the broad bandwidth of the incident radiation, and also to reduce the effects of group velocity dispersion (the generated pulse falling behind the generating pulse), one must use a short crystal. On the other hand, long crystal lengths are desired for efficient frequency conversion.

The new nonlinear crystal $\beta - BaB_2O_4$ developed by Chen^{[6.3][6.6]} provides several excellent optical properties; transparency extended from 190 nm to 2500 nm, large nonlinear coefficient, large birefringence and high damage threshold power. It allows angle-tuned phase matching over its entire transmission frequency range with high conversion efficiency in the second harmonic generation.

One important property of nonlinear crystals is the effect of group-velocity dispersion, which seriously affects the efficiency of SHG and the time resolution of pulse measurements with the SHG correlation technique for ultrashort pulses. The group-velocity mismatch is defined as

$$\frac{1}{\Delta v_g} = \frac{dk_s}{d\omega_s} - \frac{dk_f}{d\omega_f} \quad (6.34)$$

where ω is the frequency and k the wavevector and the suffices s and f denote the second harmonic and the fundamental waves respectively. The group velocity is defined

as $v_g = \frac{d\omega}{dk}$, and v_g could also be expressed as

$$v_g \approx \frac{c}{n} \left[1 + \frac{\lambda}{n} \frac{dn}{d\lambda} \right] \quad (6.35)$$

To calculate the group velocity mismatch we start with

$$\frac{dk_s}{d\omega_s} = \frac{1}{\frac{d\omega_s}{dk_s}} = \frac{1}{v_g^s} = \frac{1}{\frac{c}{n} \left[1 + \frac{\lambda}{n} \frac{dn}{d\lambda} \right]} \quad (6.36)$$

For type 1 phase matching at 497.2 nm, the s is the extraordinary ray, so using $n_e^2(\lambda)$ in equation (6.29), and Table 6.3, we can have $\frac{dn_e(248.6)}{d\lambda} = -0.9305$. And $\frac{dk_s}{d\omega_s} = 1.8958 \times \frac{1}{c}$. Similarly, using $n_o^2(\lambda)$ in equation (6.29), and Table 6.3, we can have $\frac{dk_f}{d\omega_f} = 1.6851 \times \frac{1}{c}$. So the group velocity mismatch in the BBO crystal for type 1 phase matching at 497.2 nm is 702 fs/mm.

Furthermore, the walk off angle P which determines the aperture length l_a and affects the efficiency is also an important parameter for efficient SHG. For the type 1 phase matching condition and negative uniaxial crystals, such as BBO, P is given by^[6.9]

$$\tan P = \frac{1}{2} (n_\omega^0)^2 [(n_{2\omega}^e)^{-2} - (n_{2\omega}^0)^{-2}] \sin 2\theta_m \quad (6.37)$$

where $(n_{2\omega}^0)$ and $(n_{2\omega}^e)$ are the ordinary and extraordinary refractive indices at the second harmonic frequency respectively. θ_m is the phase matching angle, which is 52.8° , at the 497.2 nm fundamental wavelength. From the data in Table 6.3, the

walk off angle can be calculated to be 3.167° or 5.53×10^{-2} rad.

Y. Ishida and T. Yajima^[6.10] have observed strong saturation of the SHG peak intensity with respect to the crystal length in their characterisation of the BBO crystal in the femtosecond region. They have found that the enhancement of the SHG peak power for a thickness of 1.9 mm relative to that for a thickness of 0.9 mm was only 20 percent, which is far less than that expected from a square law dependence.

Comly et al^[6.11] have shown by calculation that the broadening and the saturation of the peak intensity of the second harmonic pulse occurs due to the phase mismatch for the frequency components of the fundamental pulse distributing over a wide spectrum. This effect is equivalent to that of the group velocity mismatch between the fundamental and the second harmonic waves, and becomes appreciable when the group delay is much larger than the fundamental pulsewidth. Under the condition that the crystal length L is larger than the characteristic length defined by $l_c = t_p / \Delta v_g^{-1}$, the harmonic and fundamental pulses will be separated by more than one pulsewidth due to group velocity delay at the exit, and there is no more overlap between them. So the crystal length L must kept less than l_c . This theoretical result may explain the results in reference [6.10].

This is the reason why in Schafer's system^[6.2], they use 0.2 mm thickness cut at 52° and have a conversion efficiency of 8 to 10 percent. From the data sheet of the BBO crystal for type 1 second harmonic generation we can see that the angular acceptance is $\theta \times L = 0.6 \text{ mrad} - \text{cm}$, the temperature acceptance is $t \times L = 55^\circ \text{C} - \text{cm}$, the spectral acceptance is $\lambda \times L = 1.1 \text{ nm} - \text{cm}$ and the walk off angle is 56 mrad (3.2°).

The BBO crystal could be a very efficient SHG crystal in the lowest UV frequency regime.

6.3 KrF amplification

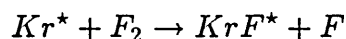
The ultrashort pulses initially generated from the stable seed oscillator at 497.2 nm are amplified by passing through the dye amplifier chain with amplified pulse energy of 50 μJ . This single amplified pulse is then frequency doubled to 248.6 nm through the BBO second harmonic generating crystal. 248.6 nm is precisely the operating wavelength of the KrF excimer amplifier.

6.3.1 The KrF amplifier

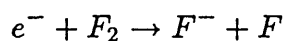
The term "excimer" refers to a molecule with a repulsive or a dissociative ground state and a bound upper state as illustrated in Fig. 6.1. The molecule excited to the upper state will relax to the bottom of the vibrational manifold and then decay radiatively to the repulsive ground state which will then rapidly dissociate. The most important type of excimer laser is the rare gas halide excimer in which the rare gas atom eg. krypton (Kr) is combined in an excited state with a halogen atom eg. fluorine (F_2).

The existence of such excimers arises from the fact that an excited rare gas atom resembles an alkali atom with an electron orbiting the core of unit positive charge. Such excited atoms are highly reactive and can form ionic bonds with the

halogens. In an electrically excited mixture of Kr and F_2 together with a buffer gas He, the Kr atoms are excited or ionised by electron impact. The excited Kr^* atoms are reactive and interact with F_2 to form KrF^*



The F_2 molecules undergo dissociative electron attachment to produce F^- ions



The positive Kr^+ ions may now recombine with F^- to produce KrF^*



where the third body M absorbs energy and momentum. This reaction occurs rapidly provided that the gas pressure is sufficiently high. Typically the Lambda Physik EMG 201 MSC excimer laser as we used, 5 percent of 80 mbar of F_2 , 120 mbar of Kr and 1.8 bar of He^[6.12].

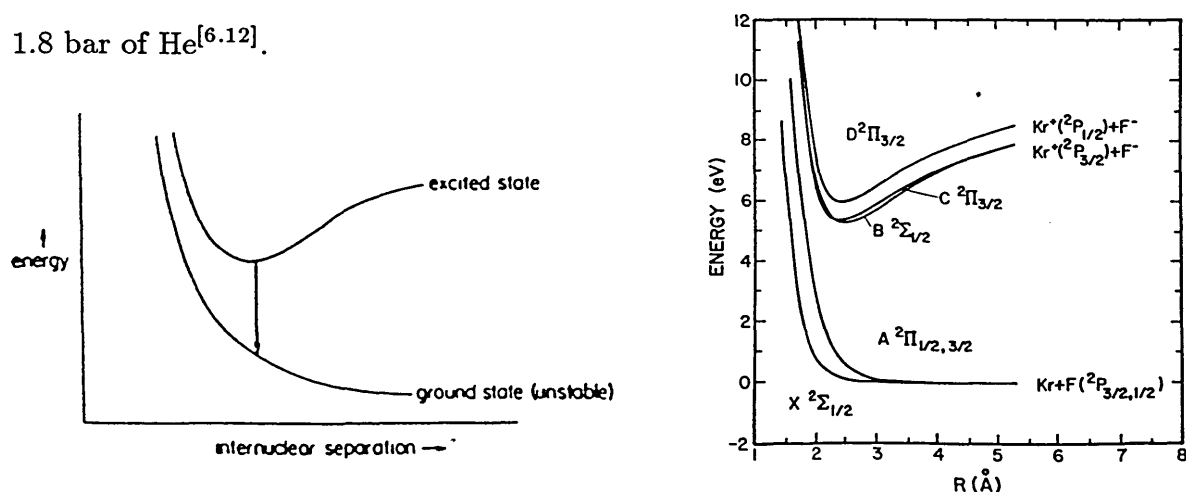


Fig. 6.1 Potential energy diagram showing the molecular structure of KrF.
[From "Excimer lasers", Editor: C. K. Rhodes]

The dissociation time of the lower level in excimer molecules is typically very short, only ~ 100 fs^[6.13]. Thus, the lower laser level is emptied very rapidly indeed, and a population difference between the lasing levels is easy to maintain. This is clearly beneficial for laser operation. Another attractive feature of the KrF rare gas halide excimer laser is its ultraviolet wavelengths as well as the tunability over a considerable wide range of wavelengths arising from the transitions available between the upper bound state and the sloping lower potential. The consequence of the repulsive nature of the ground state of the KrF excimer is the broad continuous bandwidth of transition, ~ 2 nm for KrF^[6.14]. As a result it is a suitable amplifier for ultrashort pulses. The large bandwidth of the KrF excimer also results in a small average stimulated cross section^[6.16]. This means that the saturation fluence of the KrF system can be relatively large, $\sim 2\text{mJ}/\text{cm}^2$ compared to $0.85\text{mJ}/\text{cm}^2$ for the XeCl system^[6.15]. The KrF excimer laser therefore is intrinsically a high energy system.

The EMG 201 MSC pre-ionised discharge-pumped KrF excimer laser can produce pulses of typically 30 ns duration, 500 mJ energy and with repetition rates of 100 Hz. The other significant advantage of the KrF excimer system is it can be scaled into large aperture gain modules and this enables high energy pulses to be amplified. For example, by using x-ray pre-ionising radiation instead of pre-ionising UV radiation much larger volumes of gas can be excited; or electron beams may be used to sustain the ionisation in a gas mixture in which a lower voltage discharge is being operated and very large scale systems with apertures of several tens of centimeters may be pumped using pulsed, high current, mega volt electron beams^{[6.16][6.17][6.18]}.

The high gain and wide bandwidth characteristics of the KrF excimer laser make it ideal for amplification of ultrashort pulses. Pulse durations of 60 fs have been reported to have been amplified in a KrF amplifier satisfactorily^[6.2]. However, in this high gain medium ASE can easily build up and suppress the signal amplification and the energy extraction efficiency. At longer wavelengths, saturable absorbers have been used extensively to provide a method of improving the contrast ratio of the amplified signal energy to the ASE background energy. These saturable absorbers saturate at high intensities, the weaker regions of the pulse are absorbed more efficiently than the peak region. Saturable absorbers are therefore ideal for suppressing the ASE prepulse and sharpening the leading edge of the main pulse. However, it was only recently that a saturable absorber for the KrF wavelength was developed^[6.19]. The derivative of the linear tricyclic compound acridine has a strong absorption band at the KrF laser wavelength, a methanol solution of acridine has an absorption peak of $6.8 \times 10^{-16} \text{ cm}^2$ at 248.5 nm. This may well turn out to be very useful.

Apart from depending on the saturable absorber, the system should have been designed carefully in the first place to minimise the effect of ASE. By operating at low values of small signal gain and using wide aperture gain modules ASE can also be suppressed to a low level^[6.20]. In excimer systems with a sequence of gain modules either by multiple passing or cascade stages, the pulse can be amplified without allowing the ASE levels to dominate the output if every effort is made to reduce the levels of ASE to the final amplifier. This is done by preventing the pulse from saturating the earlier stage(s) of the amplifier and by spatial filtering of the beam before each stage^{[6.21][6.22]}. Since the incoherent ASE contains higher spatial

frequencies then the amplified seed signal pulse, spatial filters or long path length between amplifier stages can help to reduce the ASE levels.

6.3.2 Amplification of ultrashort pulses in the KrF amplifier

The incoherent rate equation approach studies of the amplification of the ultrashort pulse in a KrF amplifier start with the standard rate equations for the two level system with upper level N_2 and lower level N_1 populations respectively:

$$\frac{\partial N_2}{\partial t} = -\gamma_2 N_2 - \frac{\sigma}{h\omega_{21}} I(N_2 - N_1) \quad (6.38)$$

$$\frac{\partial N_1}{\partial t} = -\gamma_1 N_1 + \frac{\sigma}{h\omega_{21}} I(N_2 - N_1) \quad (6.39)$$

and the plane-wave equation for the intensity is

$$\left[\frac{\partial}{\partial z} + \frac{n}{c} \frac{\partial}{\partial t} \right] I = gI = \sigma(N_2 - N_1)I \quad (6.40)$$

where σ is the stimulated cross section, g is the gain factor and $g = \sigma(N_2 - N_1)$, ω_{21} is the frequency difference between the upper and lower levels and γ_i^{-1} is the population lifetime of the i th state. In our sub-picosecond ultrashort pulse case compared to the ~ 1 ns KrF population lifetime, we can neglect the decay terms in equations (6.38) and (6.39). Using the population difference $N = N_2 - N_1$, those two equations may be combined as

$$\frac{\partial N}{\partial t} = -\frac{2\sigma}{h\omega_{21}}NI \quad (6.41)$$

Integrating both sides of equation (6.40) with respect to time as

$$\int_{-\infty}^{\infty} \left[\frac{\partial}{\partial z} + \frac{n}{c} \frac{\partial}{\partial t} \right] I dt = \int_{-\infty}^{\infty} \sigma NI dt \quad (6.42)$$

where $t = -\infty$ and $t = +\infty$ denote times long before and after the pulse has appeared at z , so that $I(z, t = -\infty) = I(z, t = +\infty) = 0$ and

$$\int_{-\infty}^{\infty} \frac{\partial I}{\partial t} dt = I(z, t = -\infty) - I(z, t = +\infty) = 0 \quad (6.43)$$

gives

$$\frac{d\phi}{dz} = \sigma \int_{-\infty}^{\infty} N(z, t) I(z, t) dt \quad (6.44)$$

where

$$\phi(z) \equiv \int_{-\infty}^{\infty} I(z, t) dt \quad (6.45)$$

is the pulse fluence with units of energy per unit area.

Equation (6.44) may be simplified by solving equation (6.41) for $N(z, t)$.

$$N(z, t) = N(z, -\infty) \exp\left(-\frac{2\sigma}{h\nu} \int_{-\infty}^t I(z, t') dt'\right) \quad (6.46)$$

where $N(z, -\infty)$ is the population inversion at z before the pulse has arrived. Thus we have

$$\begin{aligned} \frac{d\phi}{dz} &= \sigma N(z, -\infty) \int_{-\infty}^{\infty} I(z, t) \exp\left(-\frac{2\sigma}{h\nu} \int_{-\infty}^t I(z, t') dt'\right) dt \\ &= -\sigma N(z, -\infty) \int_{-\infty}^{\infty} \frac{h\nu}{2\sigma} \frac{\partial}{\partial t} \left[\exp\left(-\frac{2\sigma}{h\nu} \int_{-\infty}^t I(z, t') dt'\right) \right] dt \\ &= -\frac{h\nu}{2} N(z, -\infty) \left[\exp\left(-\frac{2\sigma}{h\nu} \int_{-\infty}^{\infty} I(z, t) dt\right) - 1 \right] \end{aligned} \quad (6.47)$$

From the definition of the fluence in equation (6.45) we can write

$$\begin{aligned} \frac{d\phi}{dz} &= \frac{h\nu}{2} N(z, -\infty) \left[1 - \exp\left(-\frac{2\sigma\phi(z)}{h\nu}\right) \right] \\ &= \frac{h\nu}{2} N(z, -\infty) \left[1 - \exp\left(-\frac{\phi(z)}{\phi_{sat}}\right) \right] \end{aligned} \quad (6.48)$$

where we have defined the saturation fluence

$$\phi_{sat} \equiv \frac{h\nu}{2\sigma} \quad (6.49)$$

This is the photon energy divided by twice the stimulated emission cross section.

The differential equation (6.48) may be written in terms of the ratio $\theta(z) = \phi(z)/\phi_{sat}$ as

$$\frac{d\theta}{dz} = g_0(1 - e^{-\theta}) \quad (6.50)$$

where we have used the fact that the small signal gain coefficient $g_0(z, -\infty) = \sigma N(z, -\infty)$. In many cases of interest the spatial variation of g_0 are small, and we can take g_0 to be a constant in the differential equation (6.50) for the fluence. In this case this equation has the solution

$$\phi_{out}(L) = \phi_{sat} \ln[1 + G_0(\exp(\frac{\phi_{in}}{\phi_{sat}}) - 1)] \quad (6.51)$$

Equation (6.51) gives the output fluence of a general input pulse as a function of the small signal gain, amplifier length and the saturation fluence of the amplifier. Alternatively,

$$G = x^{-1} \ln[1 + G_0(e^x - 1)] \quad (6.52)$$

where $G = \phi(L)/\phi_{in}$, and $x = \phi_{in}/\phi_{sat}$. $\phi(L)$ is the output fluence of an amplifier of length L with small signal total gain $G_0 = e^{g_0 L}$, with input fluence $\phi_{in} = \phi(0)$.

This is the standard Frantz-Nodvik result^[6.23]. It is important to note that this solution is independent of the shape of the input pulse and its reshaping during propagation in the amplifier, provided the pulse is confined to a duration which is short compared with any pumping or relaxation times.

In the limit, $x \ll 1$, that is $\phi_{in} \ll \phi_{sat}$, equation (6.52) becomes

$$G \simeq x^{-1} \ln(1 + G_0 x) \quad (6.53)$$

and if furthermore $G_0 x \ll 1$, then $\ln(1 + G_0 x) \simeq G_0 x$ and we have the small signal limit on the total gain

$$G \simeq G_0 = e^{g_0 L} \quad (6.54)$$

This limit applies to a pulse of low energy since then the growth rate follows an exponential law. However, in general it is necessary to take account of the fact that the tail of the pulse sees a different population which has in part been depleted by the leading part of the pulse and this must be described by a nonlinear time dependent gain factor.

On the other hand, for a very intense pulse, $e^x \gg 1$, that is $e^{(\frac{\phi_{in}}{\phi_{sat}})} \gg 1$, and then

$$G \approx x^{-1} \ln[G_0 e^x] = x^{-1} \ln(\exp(g_0 L + x)) = x^{-1}(g_0 L + x) = 1 + \frac{g_0 L}{x} \quad (6.55)$$

or

$$\phi_{out} \approx \phi_{in} + (g_0 \phi_{sat}) L \quad (6.56)$$

This result identifies $g_0 \phi_{sat}$ as the largest energy per unit volume that can be

extracted from the amplifier when ϕ_{in} is large compared with ϕ_{sat} . Using the fact that $g_0 = \sigma N$ and $\phi_{sat} = \frac{h\nu}{2\sigma}$, this equation could also be written as

$$\phi_{out} \approx \phi_{in} + \frac{Nh\nu}{2}L \quad (6.57)$$

This says that the largest extractable energy density corresponds to taking on average half the photons from all the excited atoms of the amplifier. The reason for the factor $\frac{1}{2}$ is simply that in the limit of large $\frac{\phi_{in}}{\phi_{sat}}$ under consideration, the amplifier is well saturated with the upper and the lower level populations being equal.

This theory of short pulse amplification is often referred to as the Frantz- Nodvik model^[6.23]. It is very useful in the design and interpretation of short pulse amplification experiments.

The process of amplification in an excimer medium can be described by several models^{[6.13][6.14][6.23][6.24]}. It is well known that the characteristic time of the dissociation of atoms in the lower state of the KrF excimer is around 50 fs^[6.13]. For long pulses, the possibility that re-excitation will occur before dissociation can be ignored and this leads to the Frantz-Nodvik model based on the simple rate equations. For pulses of ≈ 1 ps duration, the Frantz-Nodvik model provides an excellent fit to the experimentally measured gains.

However, for pulses of a few hundred femtoseconds and less, more sophisticated methods are required. Milonni, Gibson and Taylor^[6.14] have developed a theoretical technique in which the ground state dissociation is modelled as a phenomenological

population decay from the lower state of a two level system described by Maxwell-Bloch equations.

The comparison of their coherent propagation model and that of the rate equation approximation of the ultrashort pulse amplification reveals several important points: (1) The difference in the two models is most pronounced for short input pulses. The differences are small for picosecond pulses but are significant for input pulses of 0.1 ps duration. Furthermore for short pulses the differences are also significant even at low input fluence. (2) The coherent propagation model predicts a slight delay of the output pulse compared with the Frantz-Nodvik model, and this is more pronounced for short pulses. This difference may be traced to the fact that in the coherent propagation model the source for the field is the induced polarisation rather than the population difference directly and before the arrival of the pulse at any point in the medium the induced polarisation is zero and a finite time is required for the polarisation to build up to a finite value in the incoming field. (3) For ultrashort pulses the coherent propagation dynamics predict substantially different pulse evolutions than the Frantz-Nodvik model.

The broad width of the amplifying transition is one of the main reason why short intense pulses of radiation can be produced in KrF amplifier. But the broad bandwidth of the ultrashort pulses will also limit the amplification. Using the gain linewidth $\Delta\nu \approx 320 \text{ cm}^{-1}$, the pulse lengths as short as $(2\pi\Delta\nu)^{-1} \approx 17 \text{ fs}$ are near the limit for pulse amplifications without substantial temporal broadening. Even the pulse length is assumed twice of this value it already discern a pulse broadening in

the coherent propagation result since a small part of the pulse bandwidth is outside the frequency range for significant amplification^[6.14]. For the cases of 17 fs pulse duration and more at 8.5 fs, there is considerable pulse broadening predicted by the coherent propagation theory as well as a substantial reduced peak intensity^[6.14].

The output fluences predicted by the two models is the same even down to the transform limited pulse duration of 17 fs. It is only in the extreme short pulse limit that the two predictions for the output fluence differ by more than a few percent. So in our system design we can quite comfortably employ the Frantz -Nodvik model as long as for the output fluence consideration only.

K. Hill, G. New and K. Burnett have presented an alternative model^{[6.24] [6.25]} in which the lower state dissociation is rigorously described in terms of the dephasing of the lower vibrational continuum states that are excited during a downward transition from the upper excited state, although with the approximation that the lower state is linear in the region of interest and that the upper state potential is simple harmonic in character. They have used this model to study the coherent pulse propagation in a KrF amplifier.

The numerical results of this coherent interaction model have shown that many recent experimental results can still be predicted by the rate equation model without the need for more comprehensive fully coherent models because coherent re-excitation is negligible. However, as the pulse intensity becomes larger, coherent effects are likely to be very important. Hill et al, have concentrated their attention on energetic saturating pulses, and have found out that as the input pulses become more intense, the

energy gain predicted by their model tends to be less than that predicted by standard rate equations. This is because the coherent Rabi oscillations leave a significant population in the excited state even though the pulse energy is well above the saturation energy.

Comparison of Hill's coherent interaction model with Milonni's simple discrete level coherent model indicates that whilst the two models agree qualitatively, there is no quantitative agreement because of the different description of the dissociative ground state. The two models both agree in the incoherent limit, that is at low intensity with no Rabi oscillations as well as in the fully coherent limit when there are very fast Rabi oscillations since the dynamics are then independent of the dissociation of the ground state^[6,24].

Although the results of the coherent interaction model of Hill et al and the discrete level coherent model of Milonni et al agree qualitatively when only one upper state is included, it is clear that when coherent effects become important or the pulses become short the effect of additional levels in the excited state of KrF cannot be ignored. The coherent interaction model of Hill et al can be extended to include further levels in the upper bound state. However, if the discrete model is extended in the same way, the results are different. This is because the discrete level coherent model does not accurately describe the dephasing of the wavepackets on the dissociative potential.

The coherent interaction model can be limited to just two upper states to predict the behaviour of pulses 100 fs long. Coherent effects will become important

in the longer pulses as they are made more energetic (approximately greater than $5mJ/cm^{-2}$) and hence more intense. However as the pulse durations become shorter, the coherent interaction model should be extended to include further levels. Indeed for KrF a pulsewidth of 35 fs extends to about three of these upper levels.

Tilleman and Jacob^[6.26] have considered the energy amplification of short pulses in the presence of distributed absorption. A short pulse is defined as a pulse duration that is much shorter than the lasing state lifetime. They show that the energy density of a short pulse cannot be amplified indefinitely in the presence of nonsaturable absorption. In an active medium having a small signal gain g_0 and a nonsaturable absorption α the maximum attainable energy density is $\frac{E_{sat}g_0}{\alpha}$, where E_{sat} is the saturation energy. High energy density is obtained at the expense of the extraction efficiency.

Amplifier design can be optimised by choosing the parameters such as small signal gain and active medium gain length appropriately. Regardless of the input energy, the pulse energy density can be amplified to a maximum value of $\frac{E_{sat}g_0}{\alpha}$. The maximum extraction efficiency is at

$$\epsilon \equiv \frac{[\int_{-\infty}^t \phi(x, t') dt']}{E_{sat}} = \ln\left(\frac{g_0}{\alpha}\right) \quad (6.58)$$

Both very small and very large energy densities result in low extraction efficiency. In order to increase the extraction efficiency, the energy density should be maintained around a value of $\frac{E_{sat}g_0}{\alpha}$. This can be done by utilising either conventional amplifier stages of increasing aperture or an expanding beam amplifier^[6.27]. The lat-

$$E_{sat} \ln\left(\frac{g_0}{\alpha}\right)$$

ter option allows the optimisation of the extraction efficiency which is advantageous in the case of large amplifiers.

6.3.3 KrF amplification results

Figure 6.2 shows the configuration of the UV amplification system. The ultra-short seed pulse after being amplified by the dye amplifier chain is frequency doubled to the UV wavelength of 248.6 nm, that is exactly the peak gain wavelength of the KrF amplifier. The UV seed pulse is directed through the KrF gas chamber at a maximum possible diagonal path, and all the UV mirrors are used near the corresponding edges to minimise the KrF amplified spontaneous emission. There is a vacuum spatial filter after the first pass of the KrF amplifier to eliminate the KrF ASE. The seed beam had also been properly expanded to obtain the best amplification and the maximum energy extraction.

The aperture of the KrF gas chamber is 25 mm by 8 mm, so the expanded beam diameter should not exceed 8 mm.

The KrF amplifier should also be synchronised to the seed oscillator source and dye amplifier chain. Figure 6.3 shows the synchronisation of the whole system. The Lambda Physik EMG 201 MSC KrF excimer amplifier is filled with the following gas mixture: 80 mbar of 5 percent F_2 in He; 120 mbar of Krypton and the buffer gas He to a total pressure of 2000 mbar.

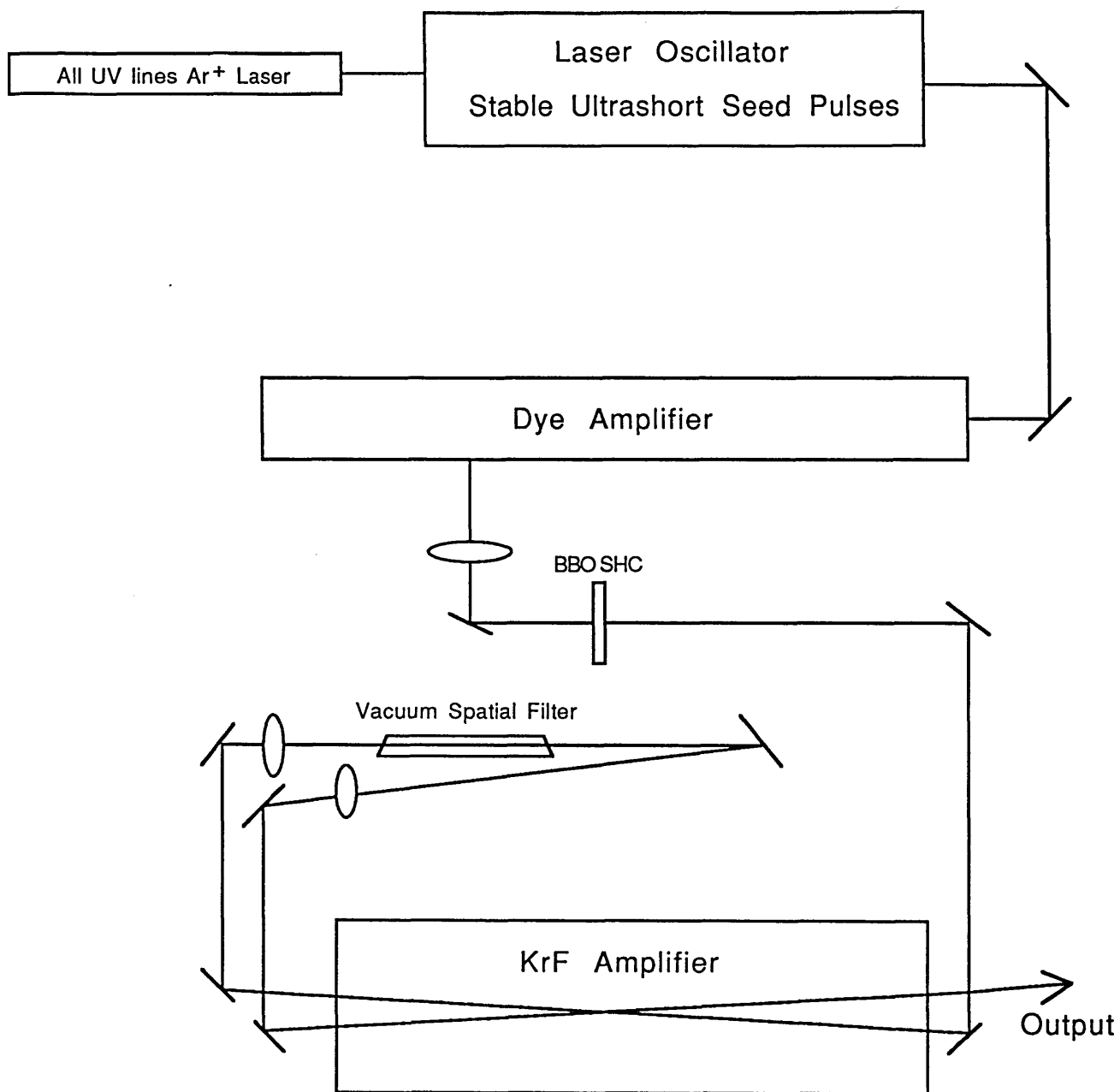


Fig. 6.2 High intensity ultrashort pulse UV laser system.

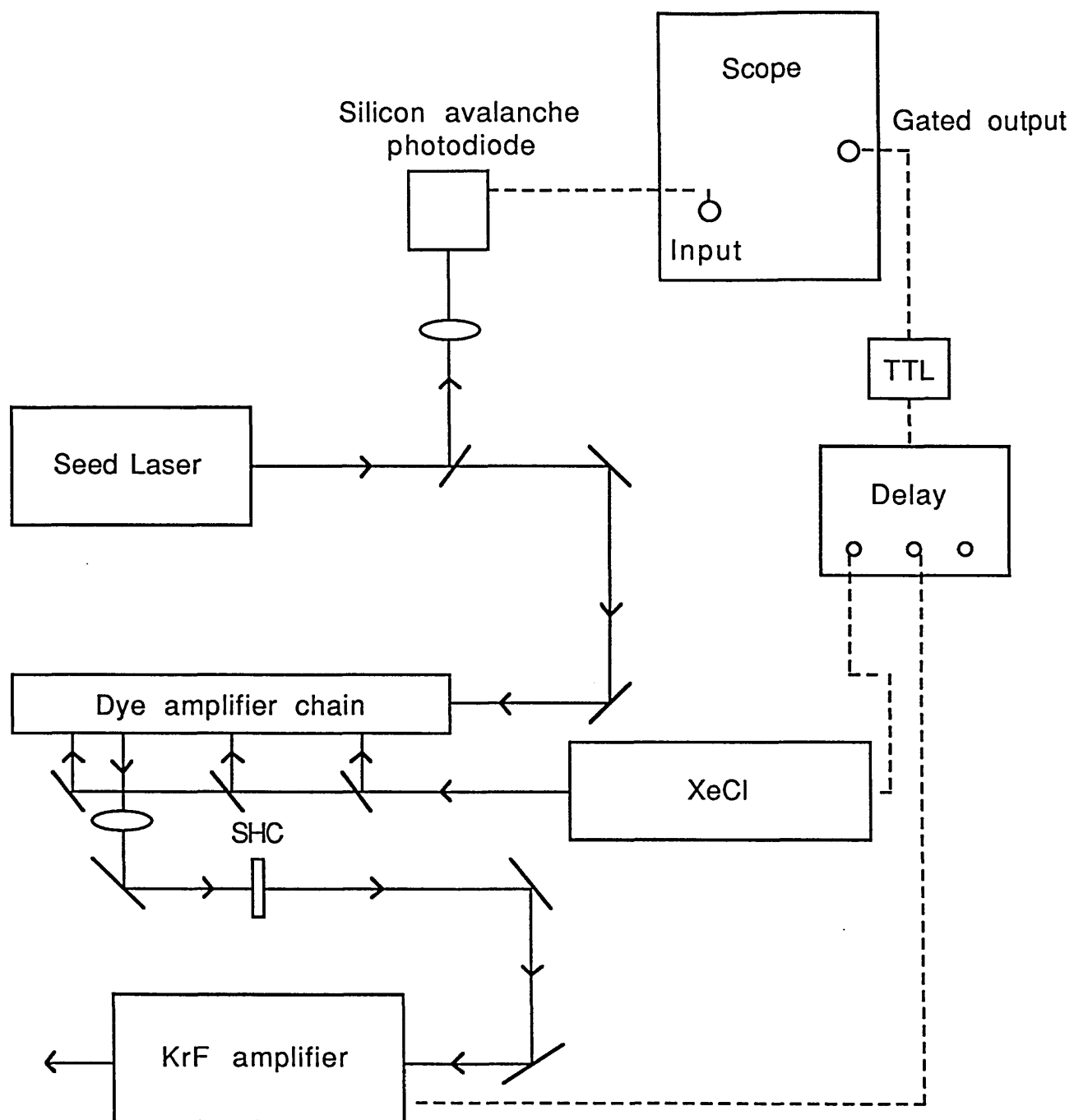


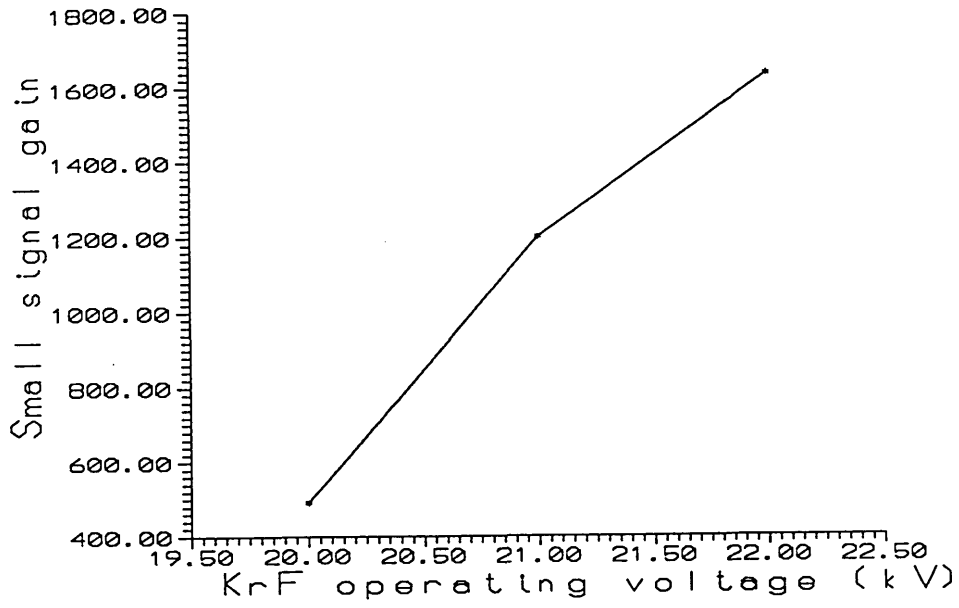
Fig. 6.3 Synchronisation of high intensity ultrashort pulse UV laser system.

The amplifier is typically operated at a working voltage of 20 to 22 kV to optimise the amplification and minimise the ASE as well as to avoid the cracks developing and resulting gas leakage in the pre-ionisation pin which can be caused by high voltages.

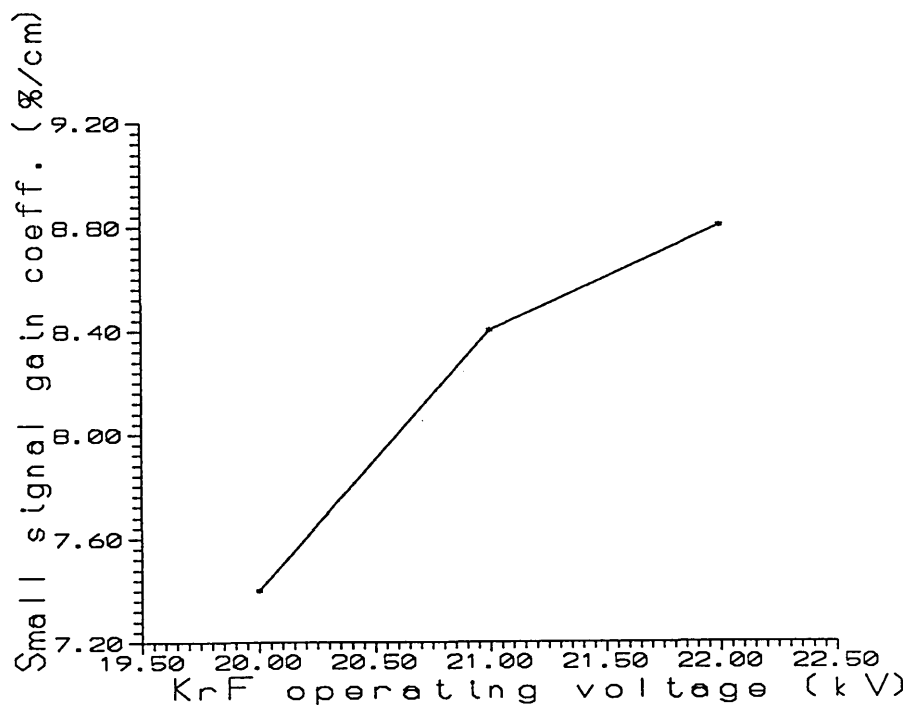
Figure 6.4 shows the small signal gain of the single pass amplification versus the KrF operating voltage, as well as the corresponding small signal gain coefficient. The maximum measured energy is 750 μJ . Since the single pass active medium gain length is 84 cm.

The input seed beam diameter is 4.2 mm, and 6 mm at the singlepass output end as well as 8 mm collimated beam after the vacuum spatial filter. Since the second harmonic generation efficiency is typically of 0.5 to 1 percent. With typical output energies of 50 μJ from the dye amplifier chain, the input fluence to the KrF amplifier can be calculated in the range between 1.76 $\mu J/cm^2$ to 3.52 $\mu J/cm^2$.

Using the small signal gain of 1633 at 22 kV operating voltage we can estimate that the output energy of 0.4 mJ to 0.8 mJ, corresponding to an output fluence in the range of 1.41 mJ/cm^2 to 2.83 mJ/cm^2 . The maximum measured single pass output energy is 750 μJ , corresponds to the output fluence of 2.68 mJ/cm^2 .



(a)



(b)

Fig. 6.4 Small signal gain (a) and small signal gain coefficient (b) of single pass KrF amplification at different operating voltage.

Using the Frantz-Nodvik relation of (6.51), we can have

$$2.68mJ/cm^2 = \phi_{sat} \ln[1 + 1633(\exp(\frac{3.52\mu J/cm^2}{\phi_{sat}}) - 1)] \quad (6.59)$$

where we have used the measured output fluence of $2.68 mJ/cm^2$, the small signal gain of 1633 and the input fluence of $3.52\mu J/cm^2$. The resultant single pass saturation fluence of the KrF amplifier at 22 kV operating voltage is $1.95 mJ/cm^2$. This is in agreement with the previously reported results of $2.0 \pm 0.2mJ/cm^2$, $2.1mJ/cm^2$ ^[6.1] and $2.1mJ/cm^2$ ^[6.28].

Figure 6.5 shows the the best fit of the Frantz-Nodvik formula using the small signal gain $G_0 = 1633$ and the saturation energy density $\phi_{sat} = 1.95mJ/cm^2$. The points are experimentally measured values. Figure 6.6 shows the typical amplified signal pulse after passing through the KrF amplifier and the vacuum spatial filter to the vacuum photodiode with calculated UV attenuators and two 250 nm narrow band pass filters in front.

Since the typical KrF discharge pulse duration is about 10 to 20 ns FWHM, and the gain depletion recovery time is of 2 to 3 ns^[6.29], we can set up a double pass configuration as shown in Figure 6.3 to obtain best amplification and maximum energy extraction.

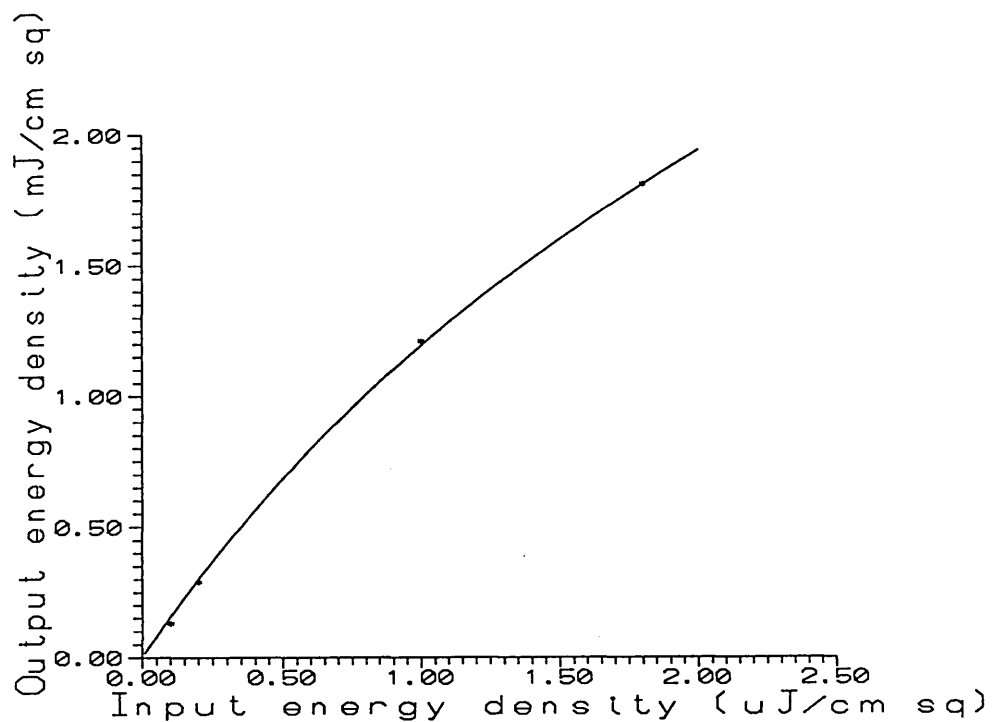


Fig. 6.5 Single pass KrF amplification results fit to the Frantz-Nodvik curve at various input energy density.

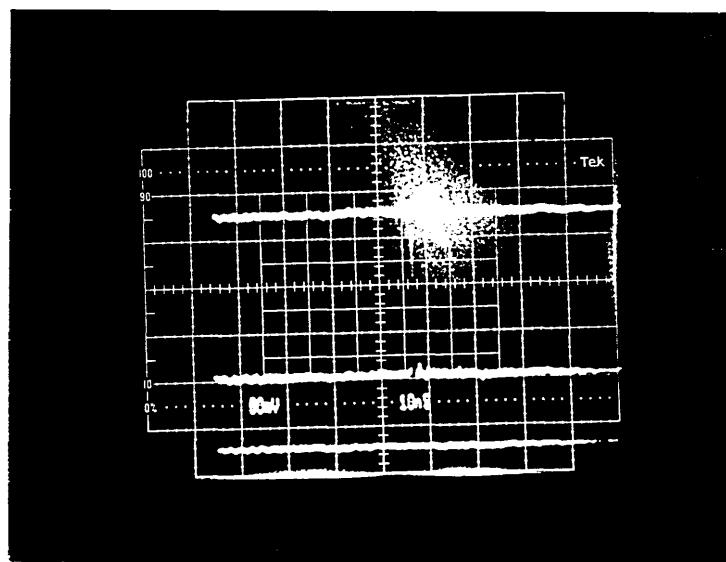
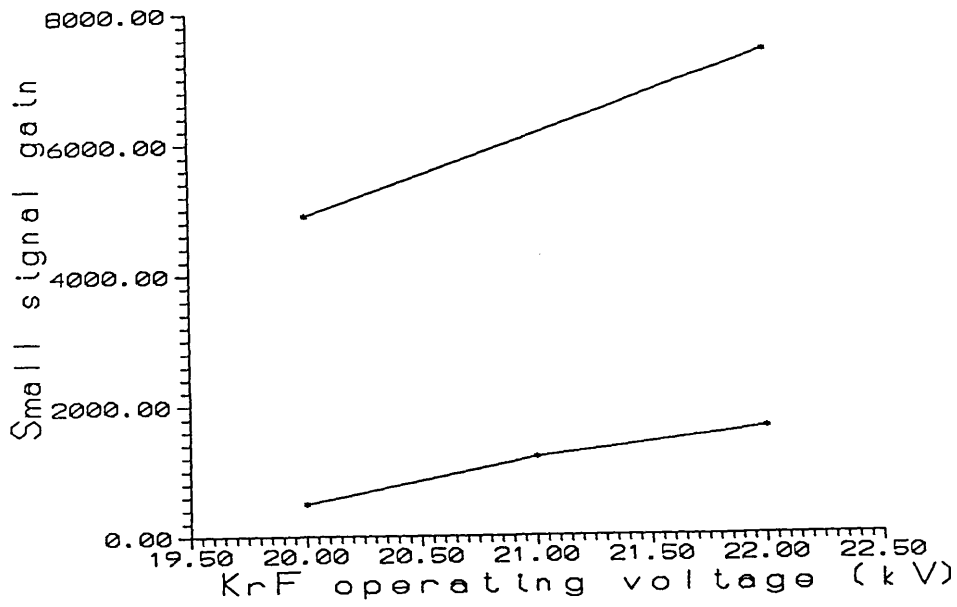


Fig. 6.6 Amplified UV pulse after passing through KrF amplifier. The bottom trace shows negligible ASE from the KrF amplifier when the UV seed beam is blocked, and the top trace shows negligible ASE from the dye amplifier chain passing the KrF amplifier.

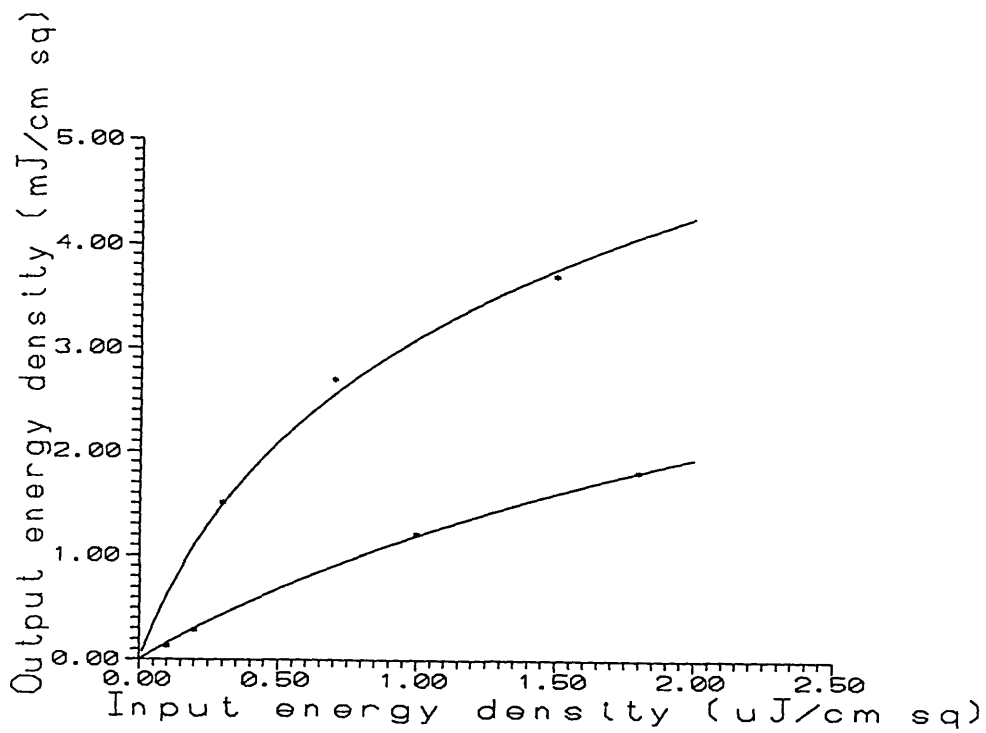
Figure 6.7(a) shows the small signal gain of the double passed KrF amplifier and the single pass amplifier for comparison and Figure 6.7(b) shows the Frantz-Nodvik curve fit. The maximum measured small signal gain G_0 is 7400 at 22 kV operating voltage and the typical measured output energy is 1.7 mJ with the output beam deflected by 3 aluminum mirrors 5 metres away from the KrF amplifier exit. Since the aperture of the EMG 201 excimer is 8 mm by 25 mm, the largest beam cross section is 8 mm in diameter. That is the effective beam area is 0.5 cm^2 . The output fluence is thus 3.4 mJ/cm^2 . Using the same input fluences and the saturation energy density of 1.95 mJ/cm^2 the Frantz-Nodvik equation gives output fluences in the range of 3.98 mJ/cm^2 to 5.19 mJ/cm^2 . This is higher than the measured fluence of 3.4 mJ/cm^2 . However, if we take into consideration the three steering aluminum mirrors of 90 percent reflectivity, then the output fluence is 4.85 mJ/cm^2 , well within the range. Figure 6.8 shows the typical KrF amplified ultrashort pulse.

6.3.4 The scaling of the KrF amplifiers

The saturation energy of the excimer stage of 2 mJ/cm^2 is rather low compared to that of the solid state lasers which are usually of the order of J/cm^2 . The output fluence could therefore very easily reach saturation as in the double or even in the single pass amplification configuration as shown in the previous section. This means that the output fluence is only linearly proportional to the active medium gain length L and so this amplification stage is really not efficient at all.

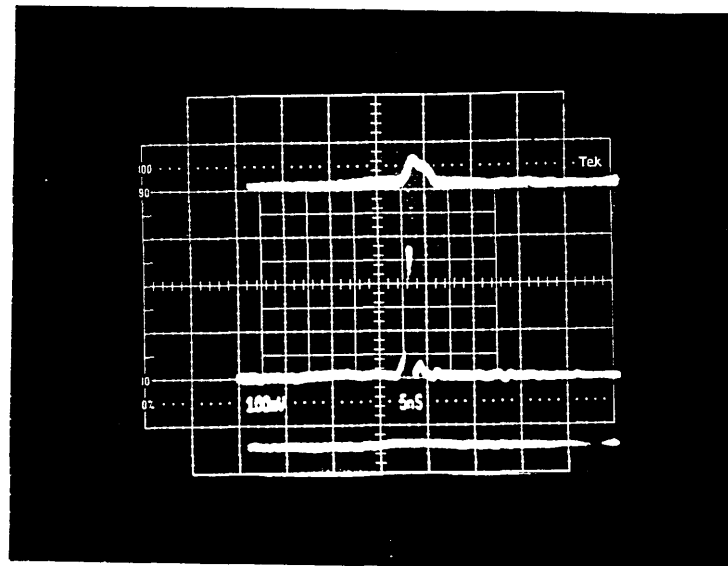


(a)

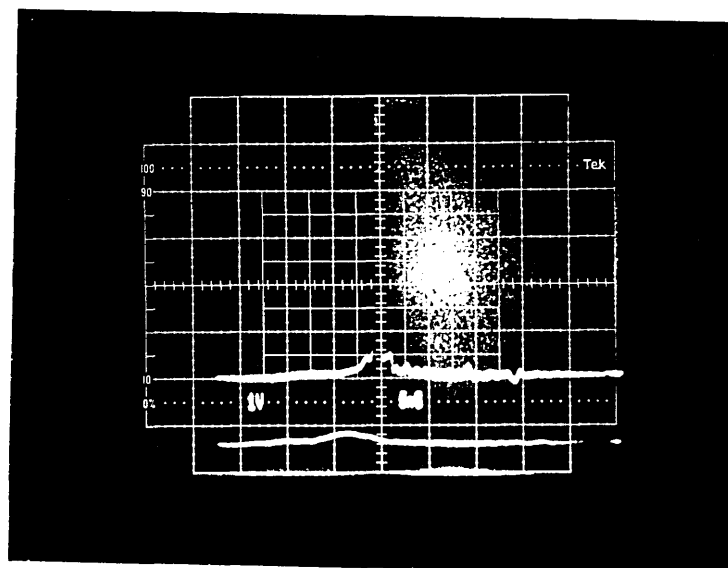


(b)

Fig. 6.7 Small signal gain of the double pass (upper trace) and single pass KrF amplification. (b) Shows the double pass (upper trace) and single pass KrF amplification results fit to the respective Frantz-Nodvik curve.



(a)



(b)

Fig. 6.8 Single pass (a) and double pass (b) KrF amplified pulse.

Although the saturation energy of the KrF amplifier is relatively low and this means that the amplification is not very efficient, as we have discussed, the system nevertheless has the advantage of scalability. We can increase the discharge cross section as in the "prometheus" system^[6.30] to up 100 mm x 100 mm cross section area which can deliver up to nearly a Joule of output energy. Also we can use the E-beam pumping instead of the discharging pumping which allows a much larger cross section with 270 mm diameter as in the "Sprite" system^[6.31] which gives out Joules of output energy. Finally, we can simply separate the seed beam into delayed orthogonal polarisation components amplified separately and then recombined to give higher output energy^[6.32].

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Chapter 7

High Intensity Ultrashort Pulse UV Laser System and Applications

We have developed a stable seed oscillator that generates ultrashort pulses of 200 fs duration at 497.2 nm. The seed pulses are then amplified by the dye amplifier chain and a single amplified pulse of 50 μJ output energy has been obtained. After frequency doubling to 248.6 nm the seed pulse is amplified by the KrF excimer amplifier to obtain a high intensity ultrashort UV pulse. This ultrashort UV source has been used to calibrate an ultrafast x-ray streak camera to the subpicosecond resolution limit. Experiments involving above threshold ionisation, high order harmonic generation, and solid target plasma interaction are in progress.

7.1 The system

Figure 7.1 shows the complete system diagram of the high intensity ultrashort pulse UV laser system. It consists of the stable seed oscillator, dye amplifier chain, second harmonic generation and the KrF amplifier.

7.1.1 Stable seed oscillator source

Ultrashort pulse duration of less than 90 fs has been generated from the

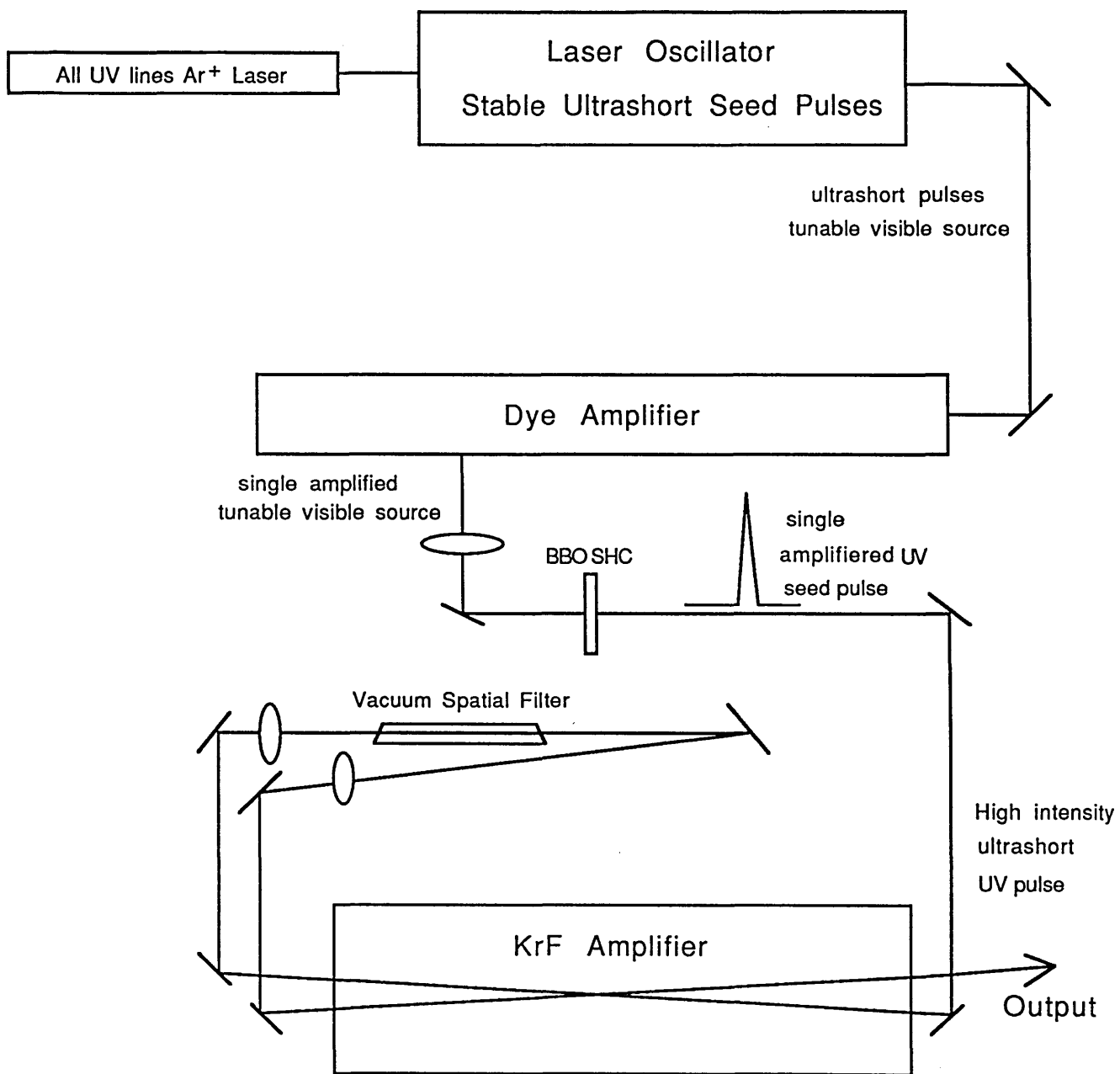


Fig. 7.1 High intensity ultrashort pulse UV laser system.

C102/DOCI CPM ring dye laser in our original development. However, for the long term operation considerations this seed oscillator source is not satisfactory because we have found that its wavelength stability is rather poor, and sometimes could even jitter out of the range of the KrF gain bandwidth which is 2 nm FWHM.

We have optimised the wavelength stabilities by isolating all the vibration sources from the circulating dye pumps, minimising the atmospheric turbulence to the saturable absorber jet since it is the most critical parameter that affects the CPM stabilities, and by using the dielectric coated tuning wedge instead of the translation slit to tune to the desired wavelength.

We have set up a linear cavity, dispersion compensated, passively mode-locked C102/DOCI dye laser that generates 200 fs pulses at 497.2 nm. The oscillator operates at 140 MHz with typical output powers of 5 to 10 mW. That is, it generates a pulse train of 35 pJ to 70 pJ energy per pulse. The seed oscillator source is tunable between 492 nm and 503 nm.

Using separate channels to monitor the seed oscillator output directly on one photodiode and the dispersed output at 497.2 nm through the spectrometer detected on another photodiode and taking the ratio of the two signals to eliminate the effect of the intensity fluctuations, we can see that any deviation is due to the wavelength jittering. Using this method, we can characterise the wavelength jitter in various time domains, and we have found that the wavelength jitter is most significant in the millisecond regime. The intensity fluctuations of the dispersive output at 497.2 nm have been calibrated to the wavelength jitter. The maximum wavelength jitter

has been optimised and characterised to be $497.2 \text{ nm} \pm 0.5 \text{ nm}$. After frequency conversion into the UV regime the wavelength jitter is $248.6 \text{ nm} \pm 0.25 \text{ nm}$, that is well within the KrF gain bandwidth.

Thus we have developed and characterised a stable seed oscillator source.

7.1.2 High repetition rate tunable ultrashort visible laser subsystem

The output energy from the seed oscillator source is only 70 pJ per pulse, and with 0.35 kW peak power. It is necessary to amplify these pulses to much higher power levels for efficient second harmonic generation. A multistage dye amplifier chain was used to boost the energy. A spatial filter after every stage is used to minimise the amplified spontaneous emission that results from the high gain of the dye amplifier. By optimising the interaction volume of the seed beam and the pumping beam we can obtain optimum amplification with minimum ASE as well as single amplified pulses. The dye amplifier chain we used is a three stage four pass system and we have optimised it to obtain best amplification with maximum energy extraction.

The typical amplification in the dye amplifier is 10^6 to 10^7 . Output energies of $50 \mu\text{J}$ to $100 \mu\text{J}$ have been obtained. The output is tunable from 492 nm to 503 nm. The pumping beam of the dye amplifier is the XeCl excimer source which can operate up to tens and hundred of hertz repetition rates.

Thus we have achieved a high repetition rate ultrashort pulse tunable visible laser subsystem.

7.1.3 μJ ultrashort UV source

The output power of the stable seed pulses after amplification in the dye amplifier chain is typically over 100 MW and with these pulses second harmonic generation is possible. A BBO crystal of 1 mm thickness cut at 52.8° is used as the second harmonic generation crystal. A conversion efficiency of 4 percent was achieved. An ultrashort UV pulse source was thus developed which could provide pulses with energies up to μJ .

7.1.4 High intensity UV source

From the very beginning our motivation in designing a suitable seed oscillator in the blue-green region and efficient second harmonic generation was to develop a KrF amplifier. We have used a Lambda Physik EMG 201 excimer modular in a double pass configuration to obtain the final amplification. The seed beam cross section must be optimised corresponding to the KrF saturation energy density of 2 mJ/cm^2 to obtain the best amplification and maximum energy extraction. A vacuum spatial filter is set up after the first pass of the KrF amplifier to eliminate the ASE.

A small signal gain of 7400 has been achieved with 1.7 mJ output energy. Using the Frantz-Nodvik formula the output fluence is in good agreement with the rate equations prediction.

The focusability of the amplified seed beam after the KrF amplifier was measured after attenuation with a focusing lens and a deconvoluted pinhole to be 1.5

times the diffraction limit. This implies that even with a typical focusing lens, the system can produce peak intensities of over 10^{16} W/cm^2 .

The main advantage of this high intensity UV system is that it can easily be scaled up. We can use cascade stages with larger and larger apertures in the usual discharge pumping scheme or in an E-beam pumped amplifier with even larger aperture. Alternatively, the UV system can be scaled up by reducing the ultrashort seed pulse duration. Pulse durations typically of 50 fs could easily be obtained from various seed oscillator sources that can either be frequency doubled to the KrF wavelength or be converted to the KrF wavelength by sum frequency mixing.

However, as the seed pulse duration is reduced down below 100 fs or so or the input fluence becomes higher, the pulse evolution in the KrF amplifier will be modified according to the coherent interaction model which we described in chapter 6.

This high intensity ultrashort pulse UV laser system is a powerful and versatile laboratory scale system. It can be used at various output stages as subsystems corresponding to the various applications. It is a self-contained system but most of the system stages could be built up separately. Furthermore, if this system is optimised still further it should be possible to use this system as the front end of an extremely high intensity system.

Figure 7.2 shows the complete high intensity ultrashort pulse UV laser system and the corresponding sub-systems available.

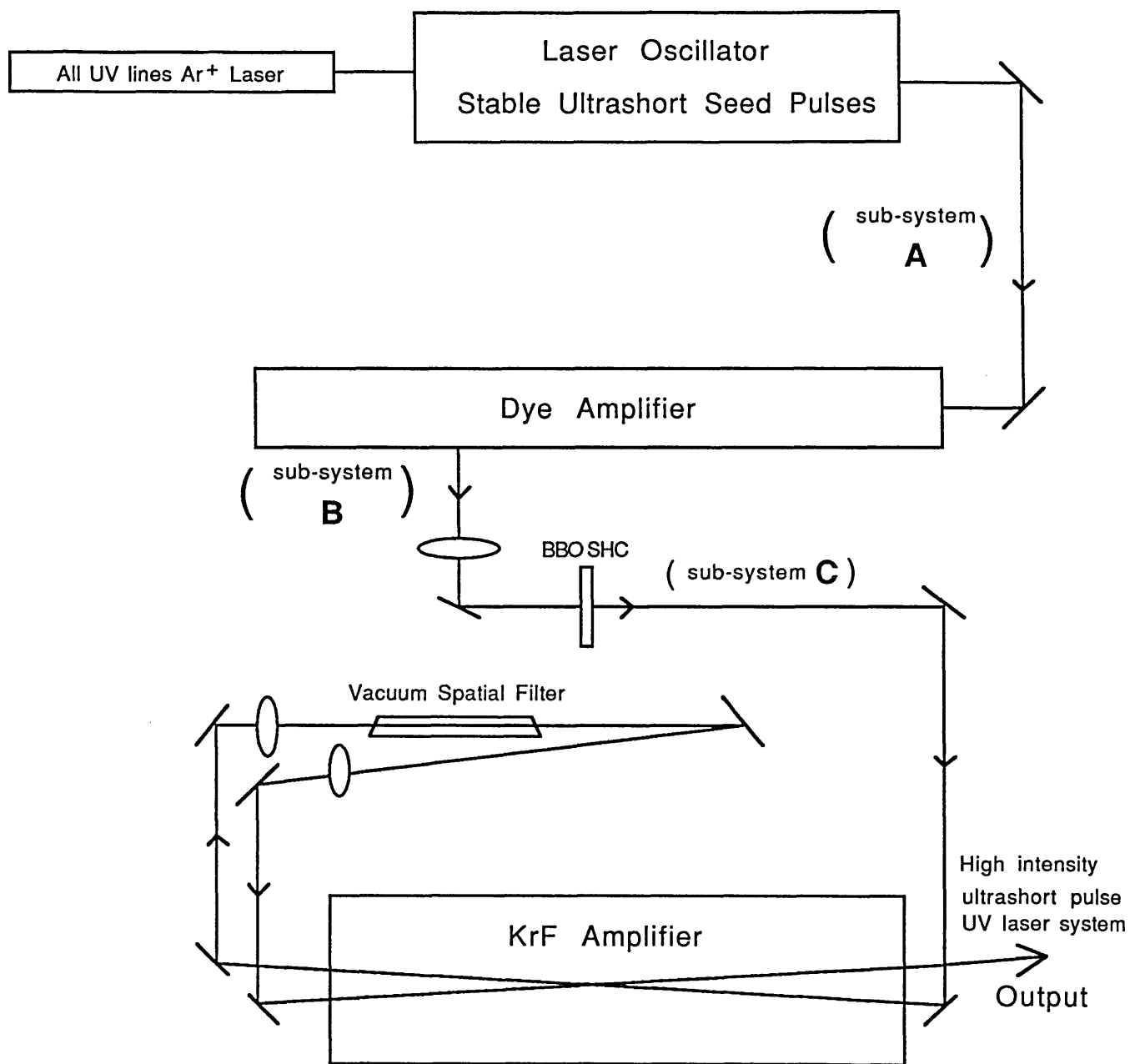


Fig. 7.2 High intensity ultrashort pulse UV laser system and sub-systems.

Sub-system **A** : Ultrashort pulses tunable visible source.

Sub-system **B** : Tunable (MW) visible source.

Sub-system **C** : Micro-joule ultrashort pulse UV source.

7.2 The applications of the high intensity ultrashort pulse UV laser system

High intensity ultrashort UV pulses could have many applications in future investigations in basic Physics. During the development of this system, the amplified UV pulse was used to calibrate the ultrafast x-ray streak camera to the best resolution reported to date. It was found that the amplified UV pulse even without any dispersion compensation had a sub-picosecond duration because it was shorter than the best resolution of 1 picosecond provided by the streak camera. We have used the amplified UV pulse to calibrate the photocathode results in a picoseconds order resolution streak camera system to verify that the time resolved result is indeed in the few tens of picoseconds region. We have also used the Imacon 500 streak camera to measure the output pulsewidth after the dye amplifier subsystem. The measured results are beyond the best resolution of 5 picoseconds, suggesting pulse duration less than 5 ps..

Experiments using the vacuum spectrometer to investigate high order harmonic generations; the electron spectrometer to measure time of flights to investigate above threshold ionisation (ATI) phenomena and the vacuum chamber to investigate the solid target plasma are under way in the Blackett Laboratory Laser Consortium.

The principal application of high intensity ultrashort laser pulses has been in the study of their interaction with atoms and molecules. It is known that an atom can absorb a large number of photons and experience multiphoton ionisation. If N photons are absorbed and the ionisation threshold is E_i , an electron can be ejected with kinetic energy $N\hbar\omega - E_i$. At high intensity, above threshold ionisation can occur

producing an electron energy spectrum given by $(N + S)\hbar\omega - E_i$, where $S = 1, 2, 3, \dots$ etc^{[7.1][7.2]}. If the laser intensity is less than $\sim 10^{13} \text{ W/cm}^2$, the heights of the ionisation peaks corresponding to different values of S decrease with increasing S in accordance with lowest order perturbation theory. However, at higher intensities the results are different owing to the fact that: (1) higher order contributions to a particular ionisation channel should be taken into account, and the perturbation theory needs to be modified thereby increasing the complexity of the calculation. (2) the interaction of the free electron with the laser field must be taken into consideration. When an electron is released, it escapes into an intense light field and oscillates with a mean energy of

$$E_{osc} = \frac{e^2 E^2}{4m\omega^2} \quad (7.1)$$

where E is the amplitude of the optical field and ω is the optical frequency. This is called the "ponderomotive" potential. The term ponderomotive refers to the "wobble" motion of charged particles in response to an applied electromagnetic field^[7.3]. So the effective ionisation potential increase to $E_i + E_{osc}$. The total energy of the released electron consists of two components, namely, the drift energy E_d equal to $\frac{1}{2}mv^2$ and the oscillation energy E_{osc} .

If the incident pulse duration is longer than the escape time from the focal region, the electron will be accelerated away from the focus centre down the gradient of the ponderomotive potential converting the energy of oscillation into increased translational energy. The ionisation peaks are therefore in the expected positions

at $E = (N + S)\hbar\omega - E_i$, except the lower ones for which $E = (N + S)\hbar\omega$ is less than $E_i + E_{osc}$. These are suppressed due to the effective elevation of the ionisation potential by E_{osc} .

However, if the pulse duration is shorter than the flight time from the focal region, the electron cannot be accelerated down the gradient of the ponderomotive potential and it is detected with the excess energy of $(N + S)\hbar\omega - E_i - U_P(r_1)$ with which it was released. This is an example of a general result: In the limit of very short pulse durations, the electron energy spectrum records the actual photoelectron energies and angular distributions at the moment of ionisation, with no final state (ponderomotive) scattering. The situation is further complicated by the fact that at high intensities, the bound states of the atom are also shifted by the high optical field, moving some states into resonance as the laser pulse sweeps past the atoms and producing further features in the photoelectron energy spectrum^[7,4].

High intensity ultrashort pulses are also finding applications in the study of the interaction of laser radiation with plasmas of solid targets. When a solid surface is irradiated by an intense laser pulse, the atoms are ionised, the electrons heated to high temperatures and x-rays are emitted. The heated plasma is ablated with a velocity of the order of 10^7 cm/sec . If it is a long laser pulse the radiation continues to interact with the expanding material up to the critical density surface beyond which it cannot propagate. Energy is therefore deposited at the critical surface and must then be conducted through the higher density plasma to the target surface. However, if the laser pulse is sufficiently short, the laser energy can be absorbed in an optical

skin depth of the solid target before significant ablation of the heated material occurs. The plasma cooling is extremely fast because of very rapid thermal conduction, energy loss to the ions, and rapid expansion of the plasma away from the site of irradiation. The fast cooling at high electron densities in turn leads to rapid recombination in the plasma thus terminating the emission of x-rays. In this way it is possible to produce intense picosecond duration pulses of x-rays. These sources will have applications in the excitation of x-ray laser systems.

As radiation becomes more intense, the response of atoms becomes noticeably nonlinear and consequently new frequencies appear in the radiation. Typically these frequencies occur at multiples of the driving laser frequency as high order harmonic generation. As long as the intensity and the order of the harmonics are not too high, the response can still be predicted by conventional perturbation theory. However this approximation fails at high intensity^[7.5]. At intensities above 10^{13} to 10^{14} W/cm^2 , the induced distortion of the atom is so great that ionisation occurs within the excitation. The induced dipole moment that accompanies this strong distortion has frequency components at odd multiples of the laser frequency. Notice that there are two sides of this problem: the response of a single atom and the collective response of an ensemble of atoms which yields strong coherent effects.

The experiments to date have been carried out at two different wavelengths, $1.06 \mu\text{m}$ and 248 nm . Both have yielded high order harmonics in noble gases: seventeen harmonics have been seen with the excimer excitation^[7.6], while up to thirty three harmonics have been reported with the Nd:glass laser^[7.7]. Therefore very inter-

esting features are expected to appear at higher intensity regimes than those used up to now. In particular, since it has been demonstrated^[7.7] that the harmonic generation at order n does not follow a simple n -power law dependence but rather saturates, we expect that increasing the intensities will produce very high odd order harmonics up to the XUV region.

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Chapter 8

Conclusions

A high intensity high repetition rate ultrashort pulse UV laser system as well as a tunable visible laser system have been developed.

An ultrashort pulse train of 200 fs pulse duration originating from the dispersion compensated passively mode-locked C102/DOCI dye laser in the blue-green region could be operated to self select a single amplified pulse in the dye amplifier chain and boost the pulse amplification up to 100 μJ output energy. The pulse, specifically tuned to a wavelength of 497.2 nm, after the frequency conversion to 248.6 nm could be further amplified in the powerful KrF excimer amplifier up to several millijoules. Even with an ordinary optical focusing system it is possible to focus the beam down to 10 to 50 μm^2 , giving a high intensity UV laser source of $10^{16} W/cm^2$.

Many immediate application in multiphoton ionisations, high order harmonic generations and ultrashort pulse solid target plasma generations could very satisfactorily employ this high intensity UV laser source.

The 200 fs pulses generated from the wavelength stabilized seed oscillator source were not optimised to generate the shortest pulse duration. Sub-hundred femtosecond pulses were generated if we removed the bandwidth limiting element of the dielectric coated tuning wedge. Further installation of pulse compression elements such as prism

pairs should enable us to optimise the output and produce shorter pulse durations.

The set up of this high intensity ultrashort pulse UV laser system has followed an efficient and straight-forward architecture. It could be operated as 'a complete high intensity UV system or it could be ~~extra~~ used at various output stages for specific applications.

In developing this system, investigations on the generation of ultrashort pulses, the characterisation of the wavelength stabilities of the seed dye laser source, the amplification in the dye amplifier chain for the best amplification, maximum energy extraction and the self selection of single amplified pulses as well as the efficient UV frequency conversion and the powerful KrF amplification have all been explored in detail. To have overcome all the practical engineering difficulties and to produce reproducible results with good stabilities are also the tasks.

In such a complex system any further research should involve close collaboration between the experimental and the theoretical aspects. A deeper understanding of the mechanisms involved would be invaluable.

The output intensity of 10^{16} W/cm^2 from this system could be scaled by employing larger aperture KrF amplifiers in cascade. Joules of output energy could be obtained^{[8.1][8.2][8.3]} and peak intensities of 10^{20} W/cm^2 should be possible. At these extremely high intensities many more exciting experimental applications could be accomplished.

The peak intensity could also be scaled by reducing the pulse durations of the

ultrashort pulses. Nowadays, pulse duration of 50 fs at specific wavelengths that can be converted to the KrF wavelength can be obtained without much difficulty. The peak intensity could be increased in this way by several times to an order of magnitude.

Apart from the well developed CPM laser which can be used to generate ultrashort pulses either at double or triple wavelengths of the KrF amplifier, there is much ~~current~~ interest in currently the high gain and broad bandwidth Ti:sapphire laser. This ^{is/} the most promising material to generate ultrashort pulses of 50 fs^[8.6], and could also be employed as an efficient power amplifier stage^[8.6].

Both the dye seed oscillator source and the Ti: sapphire solid state seed source are tunable over a wide range and could easily find suitable amplifier stages to boost the amplified seed pulse energy to a very large extent. This ensures that medium to high intensity tunable visible and near IR regime sources are available. This sub-system would also have a lot of practical applications.

The theoretical investigations of the extremely short pulse propagation in the KrF amplifier have already been well developed and published^{[8.4][8.5]}. The coherent interaction models have already predicted that the shortest pulse that could be amplified in the KrF amplifier is of the order of 17 fs. To realise this result experimentally more effort is needed in the generation and propagation of the ultrashort seed pulses, and extreme care must be taken in dealing with the temporal broadening of the ultrashort seed pulse propagating in the amplifier chain as well as in any optical materials. Since the optical length required to double the pulsewidth by

group velocity dispersion is inversely proportional to the square of the pulse duration. Furthermore, additional pulse compression techniques would need to be applied.

The system developed in this thesis has many practical applications. It is a very powerful system with a very versatile set of sub-systems. The most promising application of the complete system is that it could be used as just the front end of an ultrahigh intensity UV laser system.

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