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Winter College on Ultrafast Phenomena

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ULTRASHORT PULSE GENERATION

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SOURCES

(1) PASSIVELY MODE LOCKED DYE LASER

Colliding pulse mode locked (CPM)
dispersion compensated system
Femtosecond pulses
Tunable 450nm - 850nm

(2) SYNCHRONOUS MODE LOCKING

Dye Lasers
Picosecond and femtosecond pulses
Tunable 400nm - 1450nm
Additive Pulse Mode Locking

(3) NONLINEAR FREQUENCY SHIFTING

Raman Effect
Particularly in fibres where the broad Raman gain band can support femtosecond pulses
Picosecond pulses Dispersion problem
Compensation, femtosecond pulses
Above 1.3μm soliton shaping
fsec pulses 1.3μm - 1.8μm

PASSIVELY MODE LOCKED DYE LASER

The large homogeneously broadened bandwidth of an organic dye infers that linear amplification of femtosecond pulses is possible

Example:
Pumped at 514 nm (Argon ion laser)
Rhodamine 6G (Rh6G) lases
560nm to 650nm

$5.36 \times 10^{14}\ s^{-1}$ to $4.61 \times 10^{14}\ s^{-1}$

The $0.75 \times 10^{14}\ s^{-1}$ bandwidth would suggest that pulses as short as **13 femtoseconds** may be sustained

HOW IS THIS ACHIEVED ?

PASSIVELY MODE LOCKED DYE LASER

Historical Development

FLASHLAMP PUMPED
Rh6G + DODCI
Schmidt & Schafer Phys Lett 26A , 558 (1968)

C.W.PUMPED
1.5 psec
Ippen et al App. Phys. Lett. 21, 348 (1972)

SUB PICOSECOND PULSES
Shank & Ippen App.Phys.Lett 24, 373 (1974)

"COLLIDING-PULSE" MODE LOCKING LINEAR CAVITY
300 fsec
Ruddock & Bradley App. Phys. Lett.29, 296 (1976)

REMOVAL OF BANDWIDTH RESTRICTIONS
120 fsec
Diels et al. Opt. Communications 25, 93 (1978)

"COLLIDING PULSE" RING CONFIGURATION

Sub 100 fsec, 65 fsec
Fork & Shank App. Phys. Lett. 38, 671 (1981)

"CHIRP" & DISPERSION CONTROL

Negative chirp due to saturation of the absorber
55 fsec
Dietel et al Optics Letters 8, 4 (1984)

DISPERSION CONTROL

Prism pair sequence - Dominance of self phase modulation in system
Fork et al. Optics Letters 9, 150,153 (1984)

INFLUENCE OF DIELECTRIC MIRROR DISPERSION ON ULTIMATE PULSE DURATION FROM LASER SYSTEMS

Svelto et al Optics Letters 9, 335 (1984)

OPTIMIZED, COLLIDING PULSE, DISPERSION COMPENSATED, PASSIVELY MODE -LOCKED DYE LASER (CPM)

Rh6G - DODCI 27 fsec
Valdmanis et al Optics Letters 10, 131 (1985)

Rh6G+Kiton Red - DODCI+Malachite Green 25fsec
Taylor et al. Optics Commun 76, 229 (1990)

NEW DYE COMBINATIONS

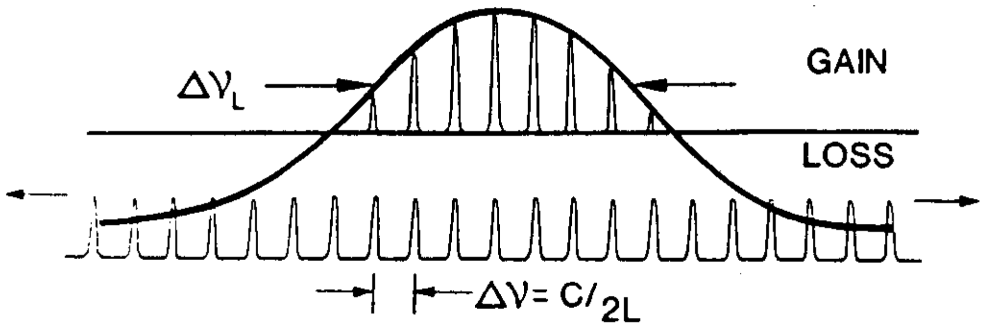
Complete visible
French & Taylor Rev Phys App 22, 1651 (1987)
Laser Focus 25, 59 (1989)

90 fs	**497nm**	**French & Taylor**	**OL 13,470**	**(1988)**
80 fs	**530nm**	**French & Taylor**	**OL 14,217**	**(1989)**
58 fs	**685nm**	**Georges et al**	**OC 69,281**	**(1989)**
36 fs	**775nm**	**Georges et al**	**OL 14,940**	**(1989)**
50 fs	**800nm**	**Georges et al**	**OL 15,446**	**(1990)**

PASSIVE MODE LOCKING

REVIEW - Has previously been introduced

CONSIDER A LASER

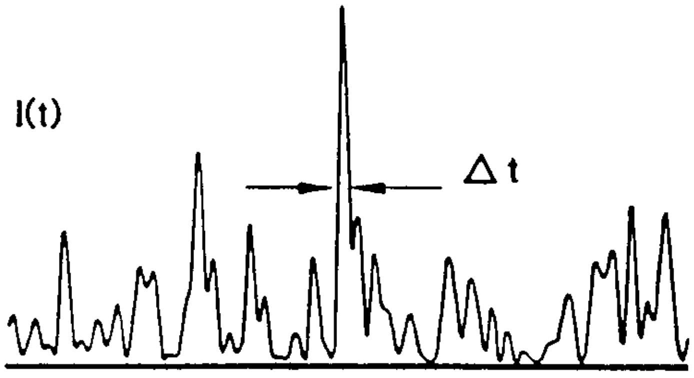


Mode spacing $\Delta\upsilon$ $= c\ /\ 2L$ $=3\text{X}10^8\ /3$
$= 100$ MHz
Round trip time $=$ 10 nsec

Typical dye laser bandwidth
$\Delta\lambda$ $=$ 5 nm at 600 nm
$\Delta\upsilon_L$ $=$ $4\ \text{X}\ 10^{12}$ Hz

Approximately $4\ \text{X}\ 10^4$ modes

When the modes have **no fixed phase relationship**, the laser output has the appearance of **NOISE**



When mode locked, a well defined single pulse propagates in the cavity, the duration of which to a good approximation is given by

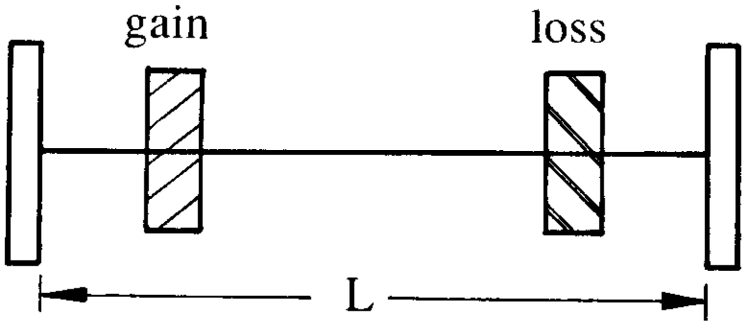
$$\tau_{RT}\ /\ \tau_{PULSE} = N$$

$$\tau_{PULSE} = (\Delta\upsilon)^{-1}$$

$$= \frac{10\ \text{X}\ 10^{-9}}{4\ \text{X}\ 10^4}$$

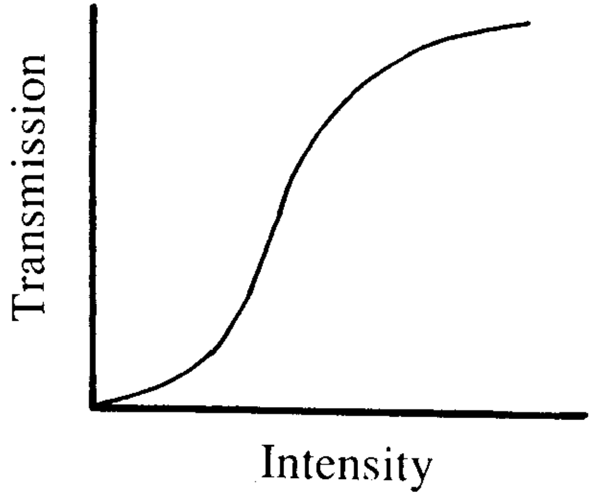
$$= 250\ \text{fsec}$$

PASSIVE MODE LOCKING



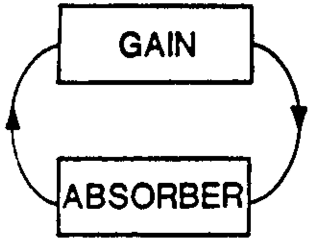
$\Delta\upsilon = c / 2L$

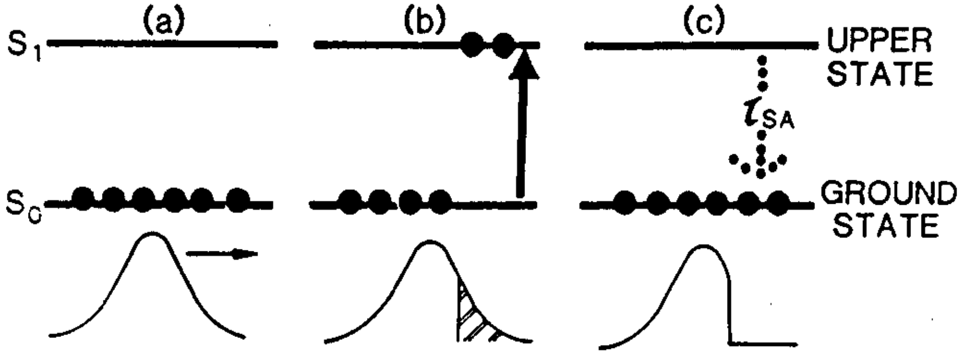
SATURABLE ABSORBER

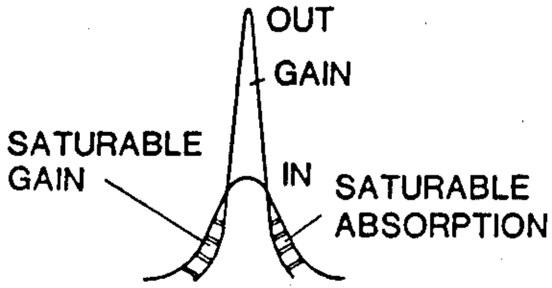


The passive mode locking mechanism

Visualize a burst of radiation in the cavity







Organic dyes have absorption (and gain) cross sections of order of 10^{-16} cm^{-2}, consequently the **saturation fluence**

$$h\upsilon / \sigma$$

is of the order of

mJ . cm^{-2}

Consequently SATURABLE GAIN and SATURABLE ABSORPTION can be readily achieved

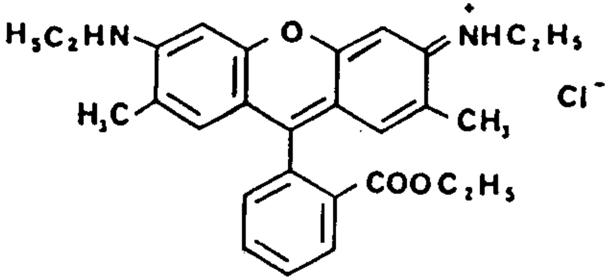
THE DYES USED
Classical System

GAIN MEDIUM Rhodamine 6G

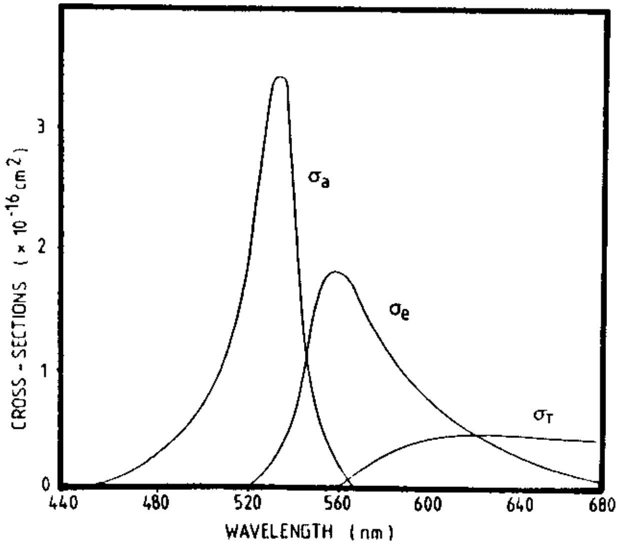
o-(6-Ethylamino-3-ethlyamino-2,7-dimethyl-3H-xanthen-9-yl) benzoic acid ethylester

SOLVENT : Ethanol / Ethylene Glycol

DYE STRUCTURE

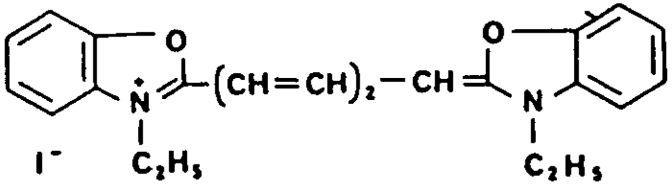


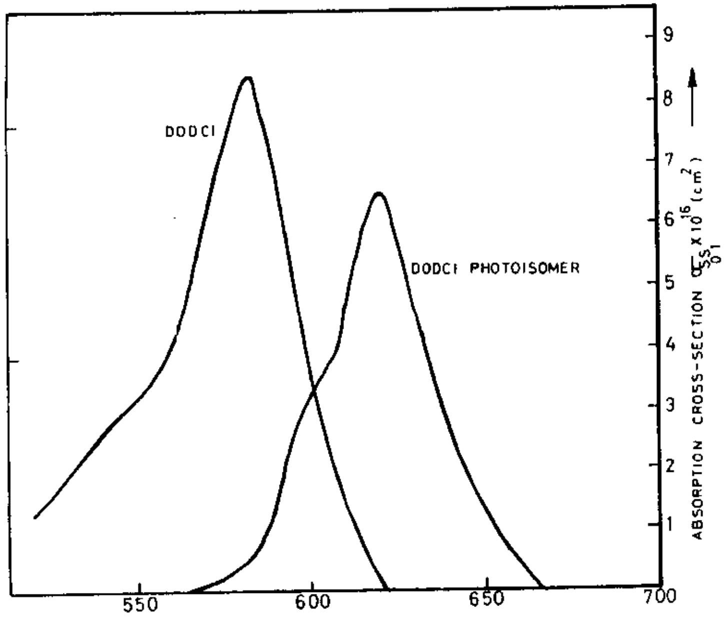
ABSORPTION / FLUORESCENCE



SATURABLE ABSORBER

3,3'-Diethyloxadicarbocyanine iodide
DODCI



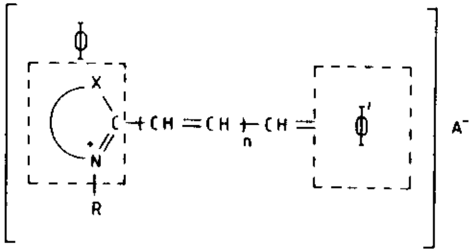


$\sigma_{ABS}(620nm) = 1 \text{ X } 10^{4}$ l/mol.cm
$(3.84 \text{ x } 10^{-17} cm^{-2})$

Recovery time τ = 1.2 nsec

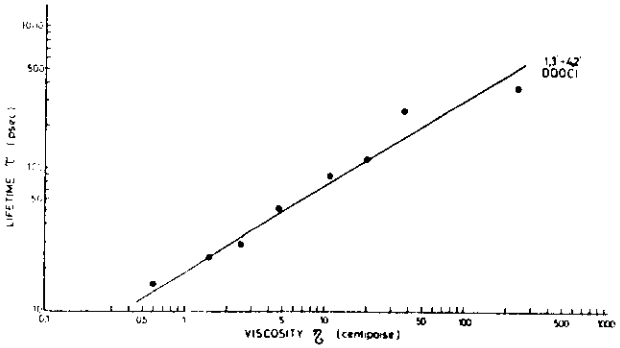
SATURABLE ABSORBERS

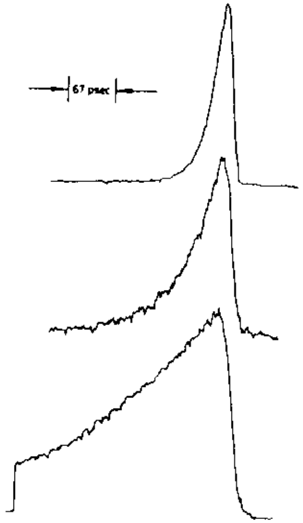
Generally dyes of the **CYANINE** family, although STYRYLS, MEROCYANINES,and XANTHENES have been successfully used.



LOOSE EXCESS ENERGY BY VIBRATION / ROTATION OF AROMATIC RINGS

THEREFORE, RECOVERY TIME CAN BE MODIFIED BY USE OF VISCOUS SOLVENTS





The pulse formation mechanism in a passively mode locked dye laser is distinct from the passive mode locking of, for example, a solid state laser

The pulse durations are shorter than the lifetime of the gain T_{1A} or that of the absorber T_{1B}.

In solidstate systems the ultimate pulse duration is determined by the recovery time of the absorber

In a dye system, for example R6G/DODCI

Recovery time T_{1B} = 1.2 nsec
Pulse durations < 100 fsec

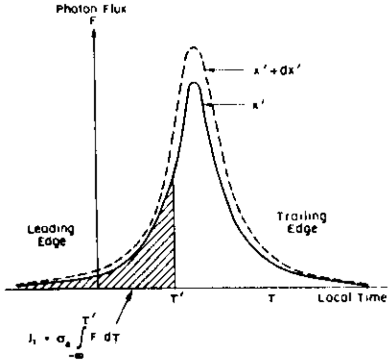
Clearly NOT limited by T_{1B}

SATURABLE AMPLIFICATION
Plays a dominant role

New (Opt. Commun 6, 188 (1972)) used a rate equation analysis to describe pulse formation in a cw dye laser

$$T_{PULSE} << T_{1A}$$
$$T_{PULSE} << T_{1B}$$

Mode locking with a **"slow" absorber**



A pulse has roughly developed from the uniform cw flux in the cavity
For mode locking to continue

PEAK should be ENHANCED relative to the background
LEADING and TRAILING edges SUPPRESSED
LOSS on **L & T** **GAIN** on **P**

Can be satisfied provided

$$T_{1A} \simeq T_{\text{Round trip}}$$

and $\sigma_B / \sigma_A \simeq 2$ (S parameter) .

$$S = \sigma_B a_A / \sigma_A a_B$$

where a is the relevant area

Having **S > 2** ensures that saturation of the absorber occurs before saturation of the amplifier

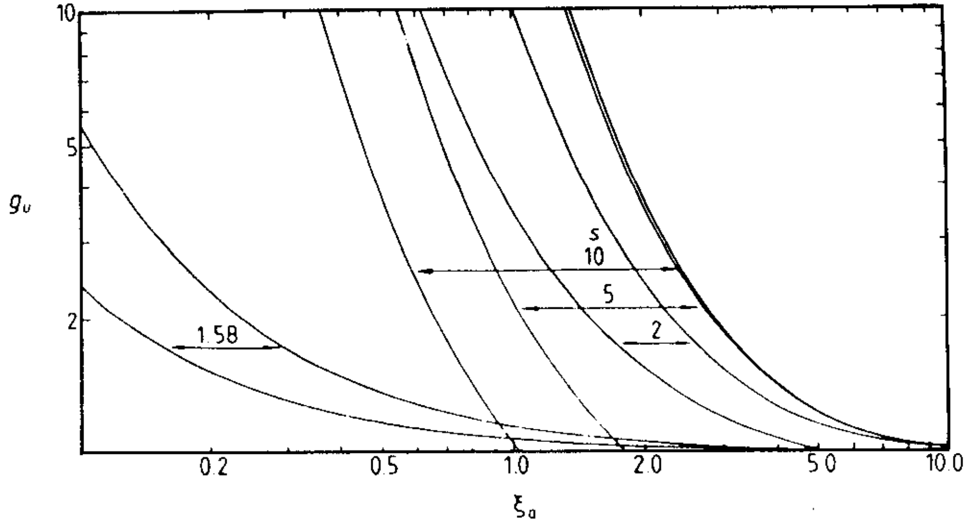
Also having

$$T_{\text{ROUND TRIP}} / T_{1A} = \zeta \simeq 1$$

ensures that the amplifier does not fully recover between transits, so that there is a net loss on the leading edge

Can all be simplified to a stability diagram

New (IEEE J.Quantum Elect. QE10, 115 (1974))



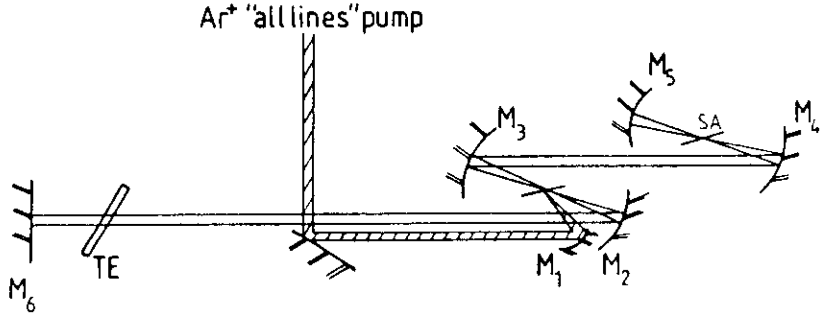
PASSIVELY MODE LOCKED CW DYE LASER

ADVANTAGES

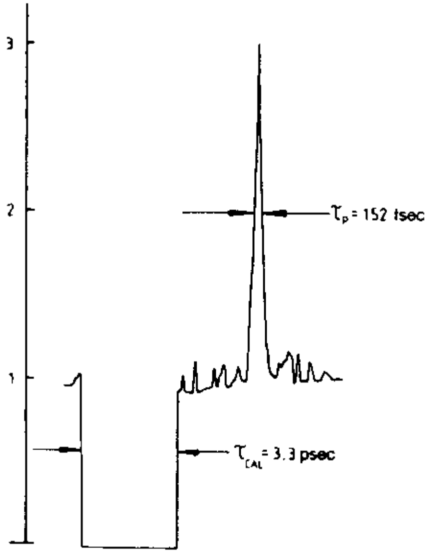
- Pulse durations < 30 fsec
- High repetition rates > 100 Mhz
- Good amplitude stability
- Low interpulse jitter < 1 psec
- Non critical cavity length
- Several pump sources
- Tunable Contrary to popular belief !

A typical cavity configuration

Linear cavity
No dispersion correction
No colliding pulse effect

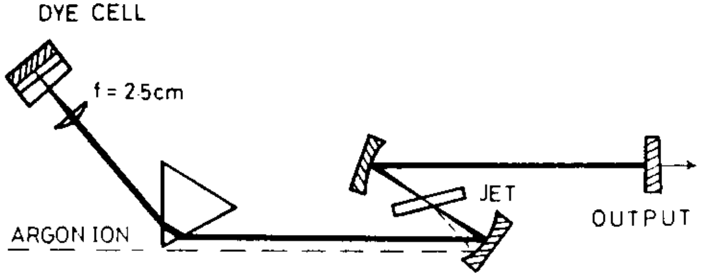


Typical pulse duration



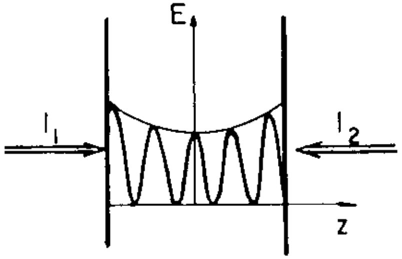
THE " **COLLIDING-PULSE** " MECHANISM

Initial idea in cw lasers
Ruddock & Bradley App.Phys.Lett. 29, 296 (1976)



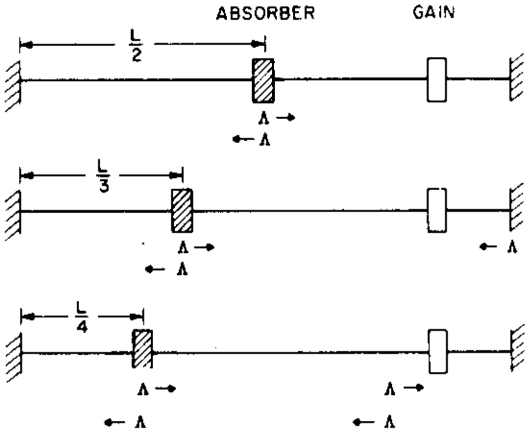
Disadvantage : High power densities at the mirror - damage

Central idea :
The collision of two pulses in the cavity enhances the effectiveness of the saturable absorption in the passive mode locking mechanism



Pulses coherent - Interfere with each other
Anti node - Intensity is greatest saturation increased and minimum loss
Node - absorber unsaturated, but field is a minimum - minimizing loss

In a linear geometry a CPM scheme can be used, without using a contacted end mirror cell



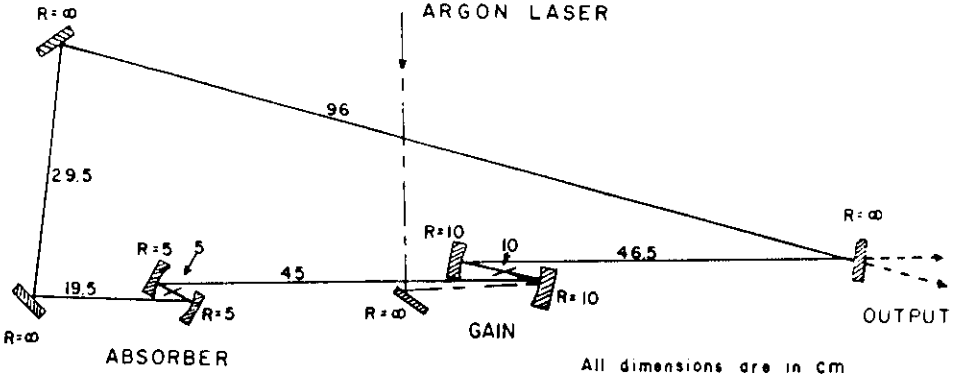
PROBLEM : Difficult to align
Precision of **position** of saturable absorber at a sub multiple of the cavity to an accuracy **of microns**

The net effect of the colliding pulse geometry is to **increase** the effective **saturation parameter** by a **factor of 3**

USE OF A RING GEOMETRY NEGATES THE ALIGNMENT PROBLEM

Mimimum energy loss takes place when collision occurs at the absorber between oppositely travelling pulses - AUTOMATIC SYNCHRONISM

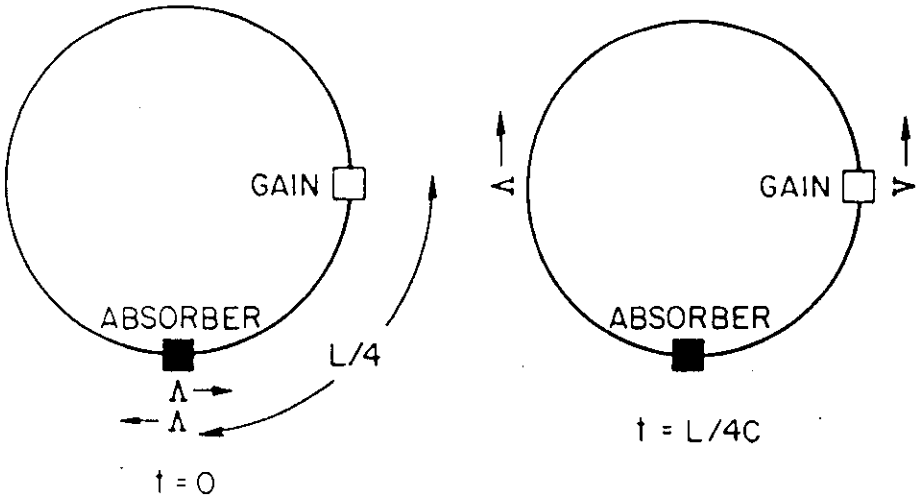
COLLIDING PULSE MODE LOCKING
Fork et al. App. Phys. Lett. 38, 671 (1981)



PROPER **RELATIVE POSITONING** OF THE **ABSORBER** AND **GAIN** JETS CAN **ENHANCE** THE **STABILITY**

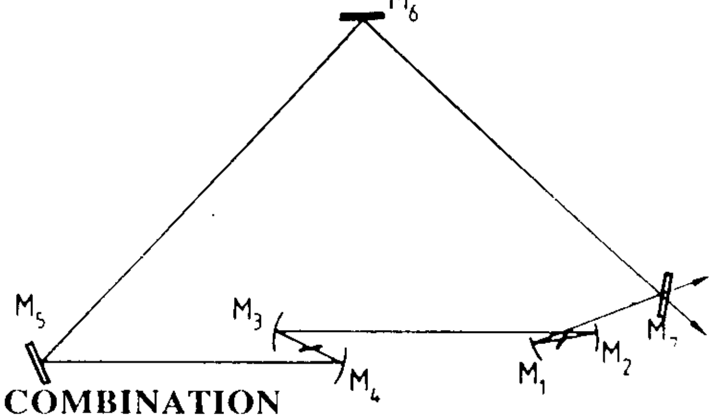
APPROXIMATELY **ONE QUARTER OF THE ROUND TRIP TIME** SEPARATION

BOTH COUNTER PROPAGATING PULSES SEE THE **SAME GAIN**



UNCOMPENSATED RING CAVITY

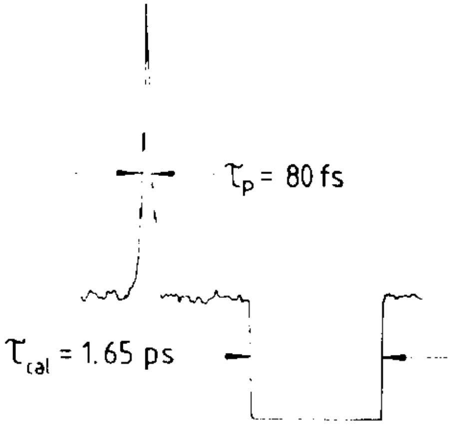
TYPICAL CAVITY ARRANGEMENT



DYE COMBINATION

Rhodamine 110
HICI (1,1,3,3,3',3'-Hexamethylindocarbocyanine iodide)
Wavelength of operation around 570 nm

TYPICAL PULSE WIDTH



IMPROVEMENTS ?

DISPERSION & PHASE MODULATION

AS PULSE DURATIONS APPROACH THE **100 femtosecond REGIME**, PULSE SHAPING IS DOMINATED BY **PHASE MODULATION AND DISPERSION**

SELF PHASE MODULATION

First treated by Shimizu
Phys Rev Lett 19, 1097 (1967)

Inherent in mode locked solid state laser systems
Arises from the **intensity dependent refractive index**

Compensation led to pulse compression
Treacy
Phys Lett 28A, 34 (1968)

Forms the basis for

Fibre - Grating Pulse Compression
Soliton shaping

SELF PHASE MODULATION IS INDUCED THROUGH THE INTENSITY DEPENDENT REFRACTIVE INDEX

$$n = n_0 + n_2 I$$

$$n_2 = 3.2 \times 10^{-20} m^2 W^{-1}$$

This gives rise to a phase shift

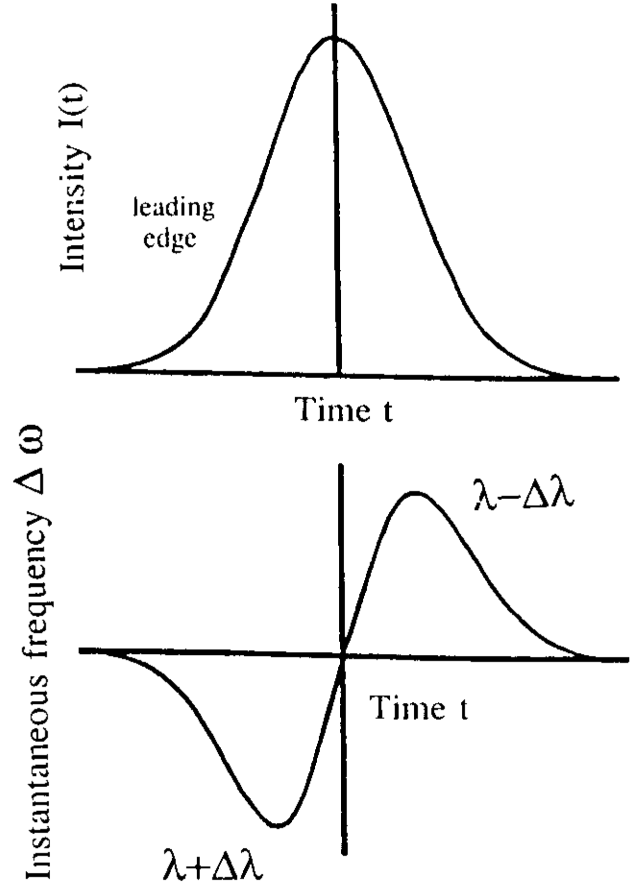
$$\Delta\Phi = dn.\,k.\,L$$
$$= n_2.\,k.\,L.\,I(t)$$

$$\Delta\omega = -d/dt\ (\Delta\Phi)$$
$$= -n_2 kL.d/dt[I(t)]$$

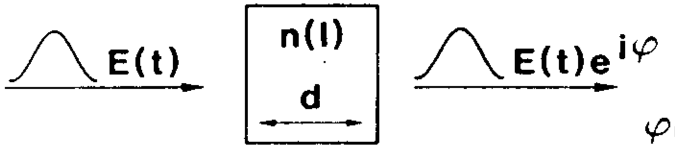
Important in the region of the beam waist of a laser resonator

FREQUENCY SHIFT α TIME DIFFERENTIAL OF PULSE PROFILE

(assuming an instantaneous response of the non linearity)

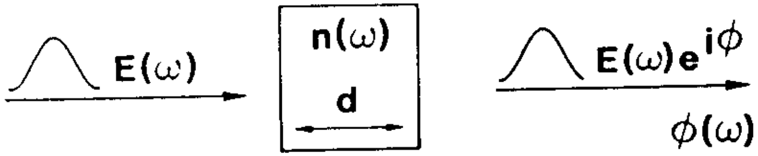


IN THE TIME DOMAIN, A TRANSPARENT MEDIUM WITH A TIME (or INTENSITY) DEPENDENT REFRACTIVE INDEX WILL EFFECT THE PHASE BUT NOT THE AMPLITUDE (shape) OF THE ELECTRIC FIELD



$$\varphi(t) = \frac{\omega n(t)}{c} \cdot d$$

IN THE FREQUENCY DOMAIN, A TRANSPARENT DISPERSIVE MEDIUM WITH A FREQUENCY DEPENDENT REFRACTIVE INDEX WILL EFFECT ONLY THE PHASE AND NOT THE INTENSITY (shape) OF THE SPECTRUM



$$\phi(\omega) = \frac{\omega n(\omega)}{c} \cdot d$$

Spectral modification

$$\Phi(\omega) = -k.d = -(2\pi/\lambda).n(\omega).\ d$$

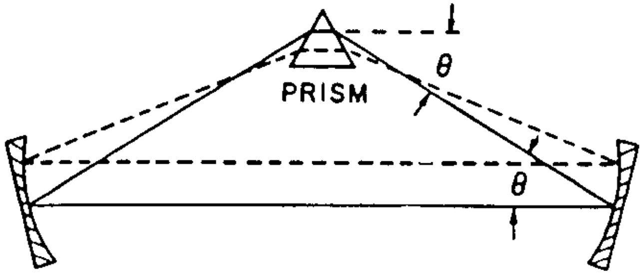
$$= -(\omega/c).n(\omega).d$$

DISPERSION

The dominant dispersion is from intra cavity elements

In a round trip the dispersion is due to

(1) Material
(2) Optical path (function of frequency)



CONSIDERING ONLY SECOND ORDER TERMS (see lecture notes of C. Brito-Cruz). The frequency dispersion

$$\phi(\omega-\omega_\ell)=\frac{1}{2c_0}(\omega-\omega_\ell)^2\left\{d_G\left[\frac{d^2(\omega n(\omega))}{d\omega^2}\right]+\left[\frac{d^2(\omega P(\omega))}{d\omega^2}\right]\right\}+\frac{1}{3!c_0}$$

where d_G is the total medium depth

For any spectral function $\chi(\omega)$

$$\frac{d^2[\omega x(\omega)]}{d\omega^2}=-\frac{\lambda^3}{2\pi c_0}\frac{d^2x}{d\lambda^2}.$$

The phase factor of the Fourier component of the light field at $\Omega = \omega-\omega_l$

$$\phi(\Omega)=-\frac{\lambda^3}{4\pi c_0^2}\left(d_G\underset{\nearrow}{\frac{d^2n}{d\lambda^2}}+\frac{d^2P}{d\lambda^2}\right)\Omega^2+$$

generally positive

Therefore **normal dispersion** can only compensate (**compress**) frequency down **shifted** (chirped) **pulses**

Intra cavity compression requires substantially thinner samples of material

Material	Index	$dn/d\lambda$	$d^2n/d^2\lambda$
BK7	1.515548	−0.03635991	0.1388040
K7	1.509825	−0.03910139	0.2031956
K10	1.500002	−0.03959791	0.1581152
BaK 1	1.570957	−0.04420354	0.2066466
F2	1.617473	−0.07357275	0.3433244
LaSF9	1.847	−0.11808	0.638166
SF10	1.724440	−0.10873030	0.5381957
Quartz	1.457	−0.030509	0.1267
ZnSe	2.589	−0.355	0.756
CaF_2	1.433	−0.0174	0.1739

Most **commonly**, the **dominant** phase modulation is **SPM** arising from the Kerr Effect in ,for example, the dye jets, giving rise to a **positive chirp**(wavelength decreasing in time)

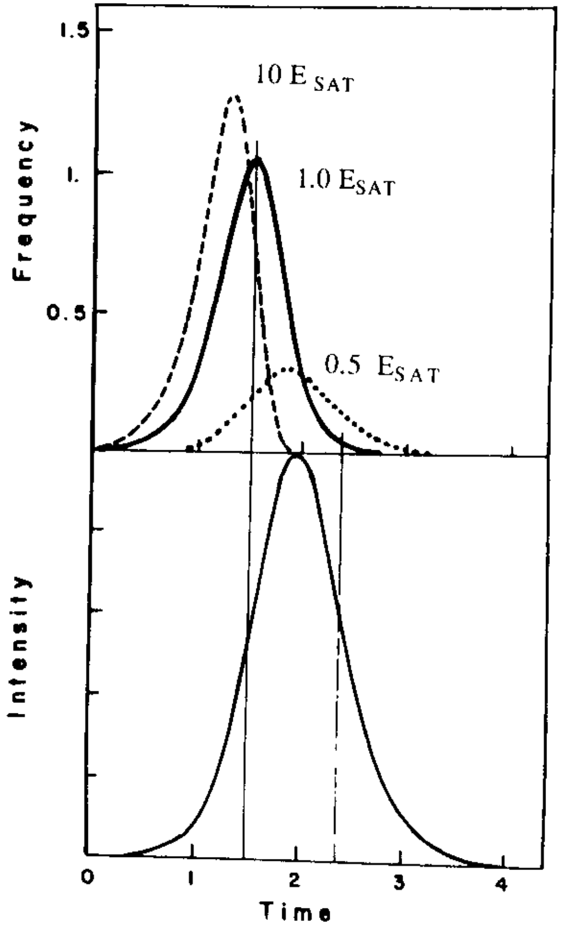
ANOMALOUS DISPERSION WILL **COMPRESS** SUCH PULSES

In the absorber dye, the operational wavelength is generally **longer** than the **absorption peak.**
As the dye **saturates**, the index of refraction will **decrease** with time
The **frequency modulation peaks** at a time such that the **accumulated energy** density equals the **saturation energy density**
If the focussing is such that the saturation energy density is reached **during the risetime** of the pulse
A uniform **down chirp** will be achieved over most of the pulse.

NORMAL DISPERSION WILL **COMPRESS** SUCH PULSES

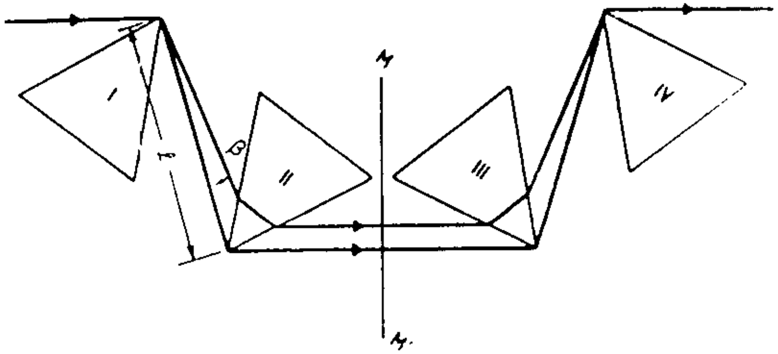
VARIATION OF SATURATION INDUCED CHIRP

Diels et al. App.Opt. 24, 1270 (1985)



THE GEOMETRIC TERM $d^2P/d\lambda^2$ CAN BE EITHER **POSITIVE OR NEGATIVE**

THE NOW CLASSIC ARRANGEMENT FOR THE INTRODUCTION OF CONTROLLABLE DISPERSION IN A CPM UTILIZES THE PRISM SEQUENCE



FOR QUARTZ

A pair separation of **$l = 250$ mm** is **equivalent** to **6.6 mm** of quartz.

ALLOWS THE EFFECT OF **DISPERSION** TO BE CORRECTED FOR AND **BALANCED** BY **NON-LINEARITY (SPM)**

LEADING TO THE CONCEPT OF **SOLITONS**

PULSE SHAPING DUE TO REFLECTION OFF DIELECTRIC MIRROR SURFACES

THE COMPLEX REFLECTION COEFFICIENT HAS A FREQUENCY DEPENDENT PHASE TERM WHICH ADDS TO THE OVERALL DISPERSION

LEADING TO **PHASE MODULATION** AND **PULSE BROADENING**

PULSE SHAPING ON REFLECTION IS A SECOND ORDER EFFECT OF THE PHASE DISPERSION BUT CAN BE EFFECTIVE FOR

$\tau<50$ fsec

Several Investigators

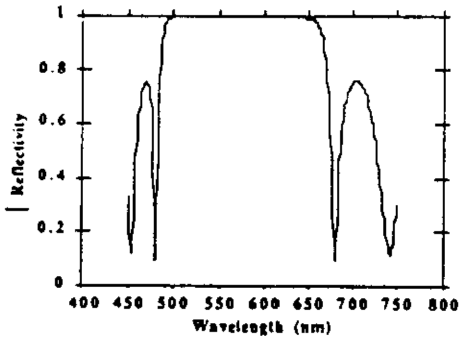
Weiner et al.	Opt.Lett. 10, 71 (1984)
Diels et al	U/fast. Phen. IV, 30 (1984)
Svelto et al	Opt.Lett. 9, 335 (1984)

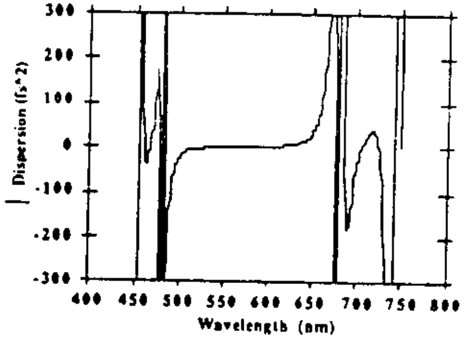
CONSIDER A STANDARD DIELECTRIC REFLECTOR

($\lambda/4$) layers, of equal thickness of the form

AIR H $(LH)^9$ SUBSTRATE

$n_L = 1.45$, $n_H = 2.28$, $\lambda = 563$ nm



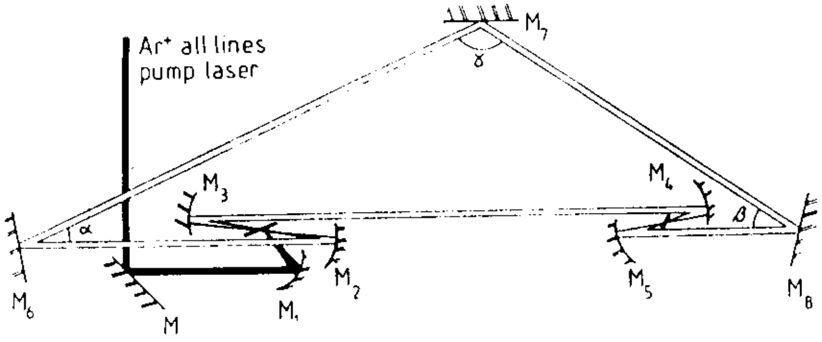


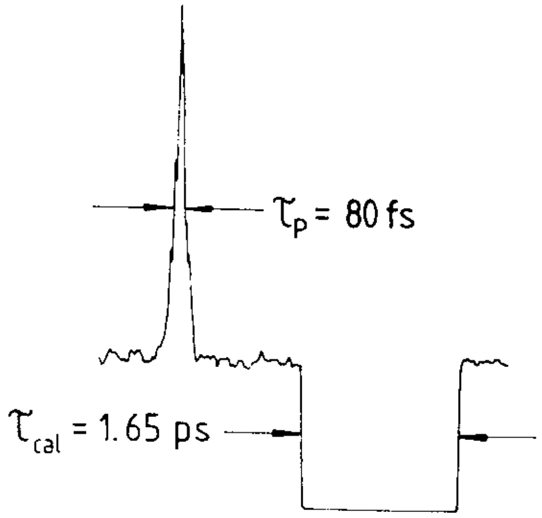
Operation at an **angle** to the mirror away from resonance gives a **long wavelength cut-off** and introduce **anomalous dispersion**

Unequal layer thickness can exhibit a large dispersive effect, even away from the band edge

CPM LASER WITH THE MIRRORS DOING THE DISPERSION CORRECTION

CORRECT CHOICE OF α, β, & γ





FACTORS CONTRIBUTING TO DISPERSIVE EFFECTS IN A CPM LASER

(1) Normal dispersion from transparent materials (glass, solvents)

(2) Mirrors

(3) Anomalous dispersion due to unsaturated loss of the absorber

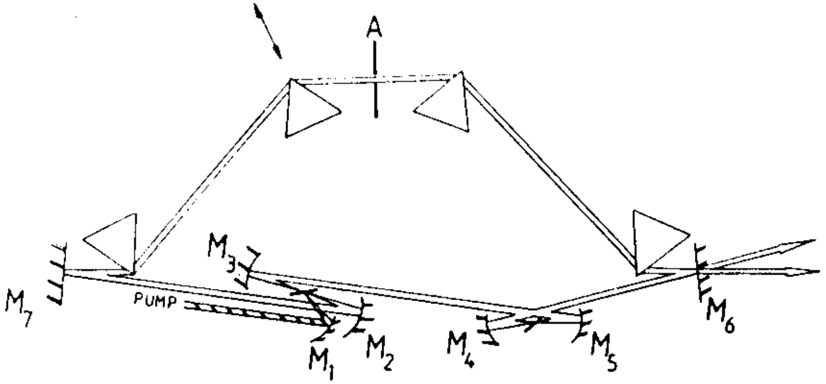
(4) Self phase modulation due to saturation of absorber

(5) Anomalous dispersion of introduced optical elements (prisms, GTI's)

Svelto et al IEEE J.Q.E. QE20, 533 (1984)

Cavity components		ϕ'' (10^{-30} s^2)	Sign of frequency chirp
Ethylene glycol jet stream (100 µm)	(λ_L = 610 nm)	- 8.4	+
Quartz (1 mm)	(" " ")	- 54	+
Flint F2 glass (1 mm)	(" " ")	- 160	+
Anomalous dispersion in DODCI	(" " ")	7.5	-
Anomalous dispersion in DODCI photoisomer	(" " ")	- 32	+
Mirror on red shifted side (4% transmission)		240	-
Self-phase modulation in DODCI	(λ_L = 610 nm)	36-300	-

THE DISPERSION-COMPENSATED, COLLIDING-PULSE, PASSIVELY-MODE-LOCKED, DYE LASER (CPM)



TYPICAL SYSTEM

Amplifier Dye	Coumarin 102
Concentration	$4X10^{-3}M$ (400 µm jet)
Passive Dye	DOCI
Concentration	$1.5X10^{-3}M$ (100 µm jet)
Round trip time	6.2 nsec
Prism separation	310 mm
Mirrors	M_2-M_7 100%R (500nm)
Output mirror	1%T
Pump power	4W (All lines UV,Ar ion)

OUTPUT

Tunable	493 nm to 505 nm
Pulse duration	< 100 fsec
Repetition rate	160 MHz
Average power	6mW (400 W peak)