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UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION
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UNITED NATIONS INDUSTRIAL DEVELOPMENT ORGANIZATION



INTERNATIONAL CENTRE FOR SCIENCE AND HIGH TECHNOLOGY

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International Centre for Theoretical Physics
Trieste, Italy

Winter College on Ultrafast Phenomena 1991

Winter College on Ultrafast Phenomena

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*Generation, Measurement, and Applications
of Ultrashort High Intensity Excimer
Laser Pulses*

GENERATION, MEASUREMENT, AND APPLICATIONS OF ULTRASHORT HIGH INTENSITY EXCIMER LASER PULSES

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1st Lecture:

Generation of Ultrashort High Intensity Excimer Laser Pulses

Abstract. Excimer lasers are the only powerful lasers in the ultraviolet covering the range from 126 nm to 352 nm. Unfortunately, they are not accessible to the usual methods for ultrashort pulse generation. They nevertheless have a sufficient bandwidth to amplify ultrashort pulses down to less than 100 fs pulse duration. This leads to the only useful scheme to first generate ultrashort pulses with dye lasers in the visible or infrared and amplify these pulses after frequency multiplication in an excimer laser amplifier.

The implementation of this scheme in the conventional method that will be briefly discussed is very complex and expensive. We thus have developed a relatively simple and inexpensive solution over the passed years which will be discussed in detail. It uses only one dual-channel commercial excimer laser as pump source for the various dye lasers stages and amplifier for the frequency-doubled input pulses, respectively, and needs no electronics at all.

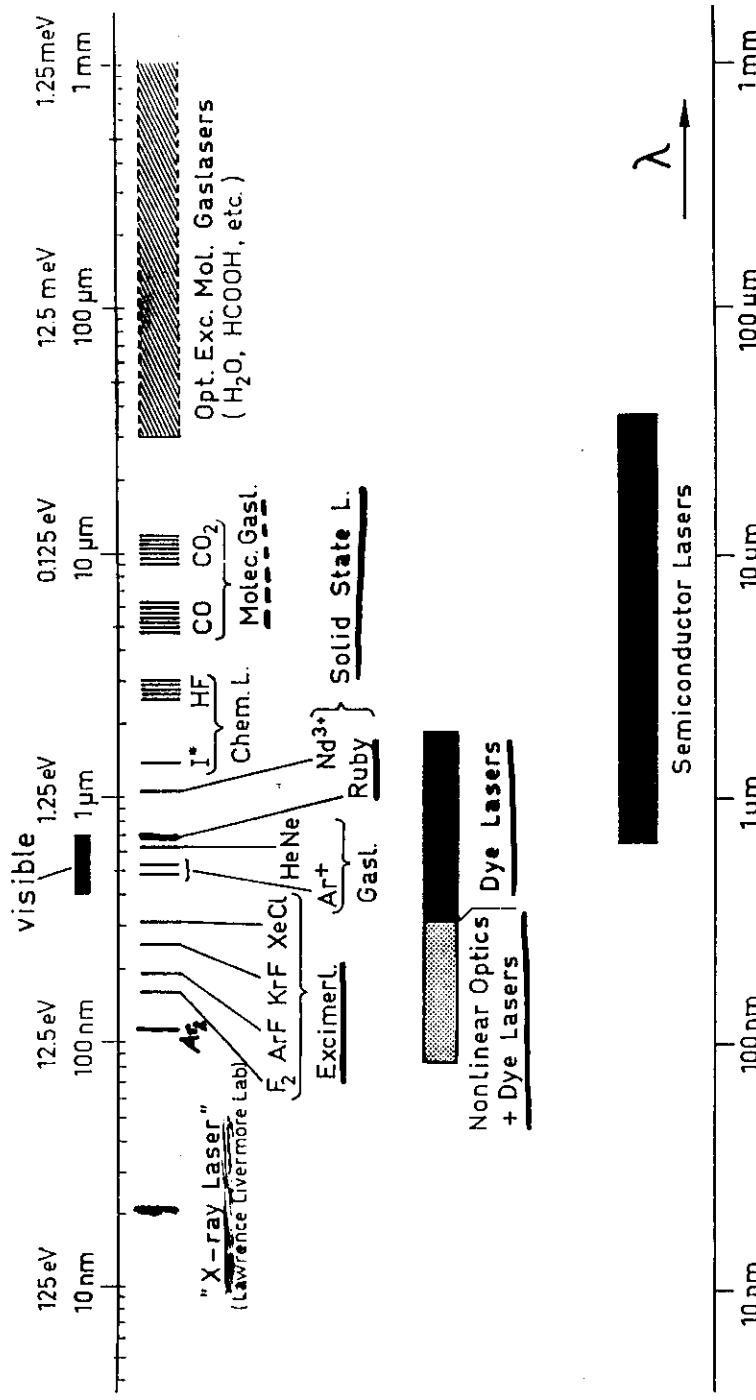
For further amplification we developed a large aperture discharge-pumped amplifier with x-ray preionization that is expected to deliver up to 300 mJ pulse energy in KrF at 248 nm. The only way to reach still higher pulse energies is then the amplification with electron-beam-pumped excimer laser amplifiers of sufficiently high cross section as will briefly be discussed in the third lecture.

1st Lecture

References

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Subpicosecond, widely tunable distributed feedback dye laser.
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Simplified laser system for the generation of 60 fs pulses at 248 nm.
Opt. Commun. 68, 196-202 (1988)
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at 248 nm.
Opt. Commun. 63, 305-309 (1987)
- 1.4 B. Dick, S. Szatmári, B. Rácz, F.P. Schäfer:
Bandwidth limited amplification of 220 fs pulses in XeCl: Theoretical and
experimental study of temporal and spectral behavior.
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- 1.5 S. Szatmári, B. Rácz, F.P. Schäfer:
Bandwidth limited amplification of 220 fs pulses in XeCl.
Opt. Commun. 62, 271-276 (1987)
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Femtosecond pulse generation at 193 nm.
ECO3, 12-13 March 1990, The Hague, The Netherlands. In: *Applications
of Ultrashort Laser Pulses in Science and Technology*. SPIE Proc. Ser.,
Vol. 1268, 22-29 (1990)
- 1.7 M. Steyer:
Discharge kinetics and emission characteristics of a large area-cold
cathode flash x-ray tube: parametric study and numerical modelling.
J. Phys. D: Appl. Phys. 23, 18-25 (1990)
- 1.8 B. Rácz, M. Steyer, H. Mizoguchi:
Gain properties of a wide aperture X-ray pre-ionized excimer amplifier.
Opt. Quant. Electron. 23, 65-72 (1991)

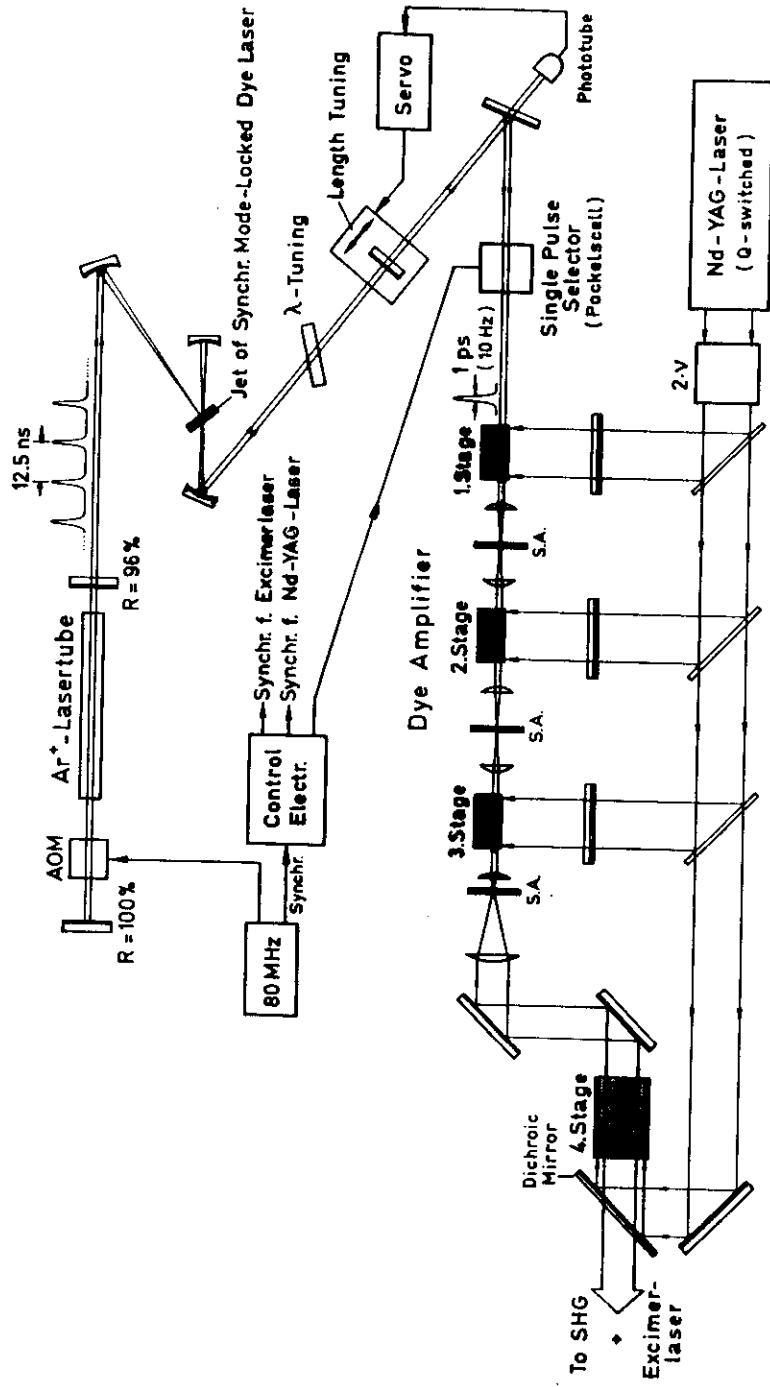
WAVELENGTH RANGE OF LASERS



N

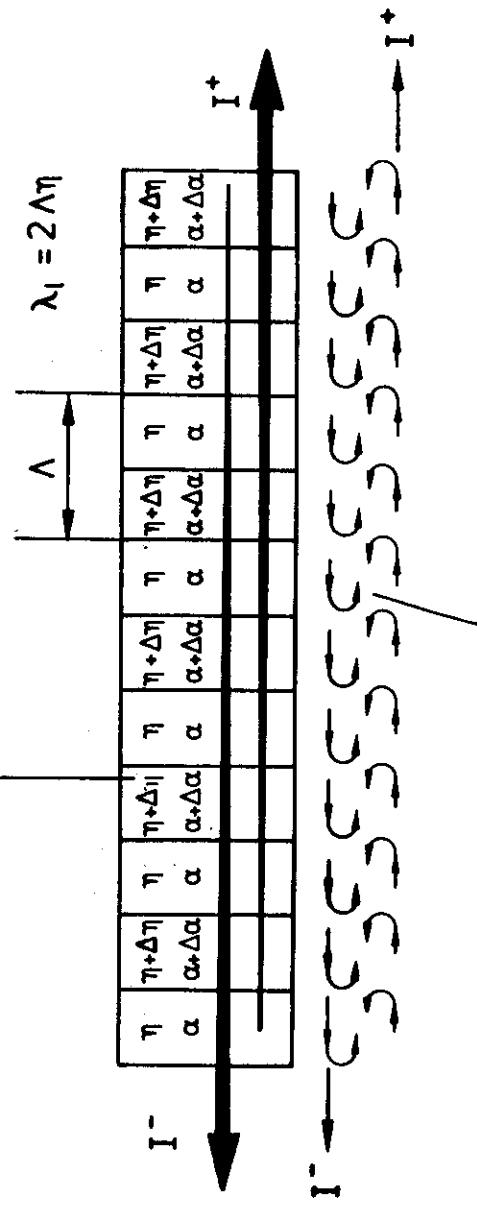
Scheme of generation of ultrashort excimer-laser pulses

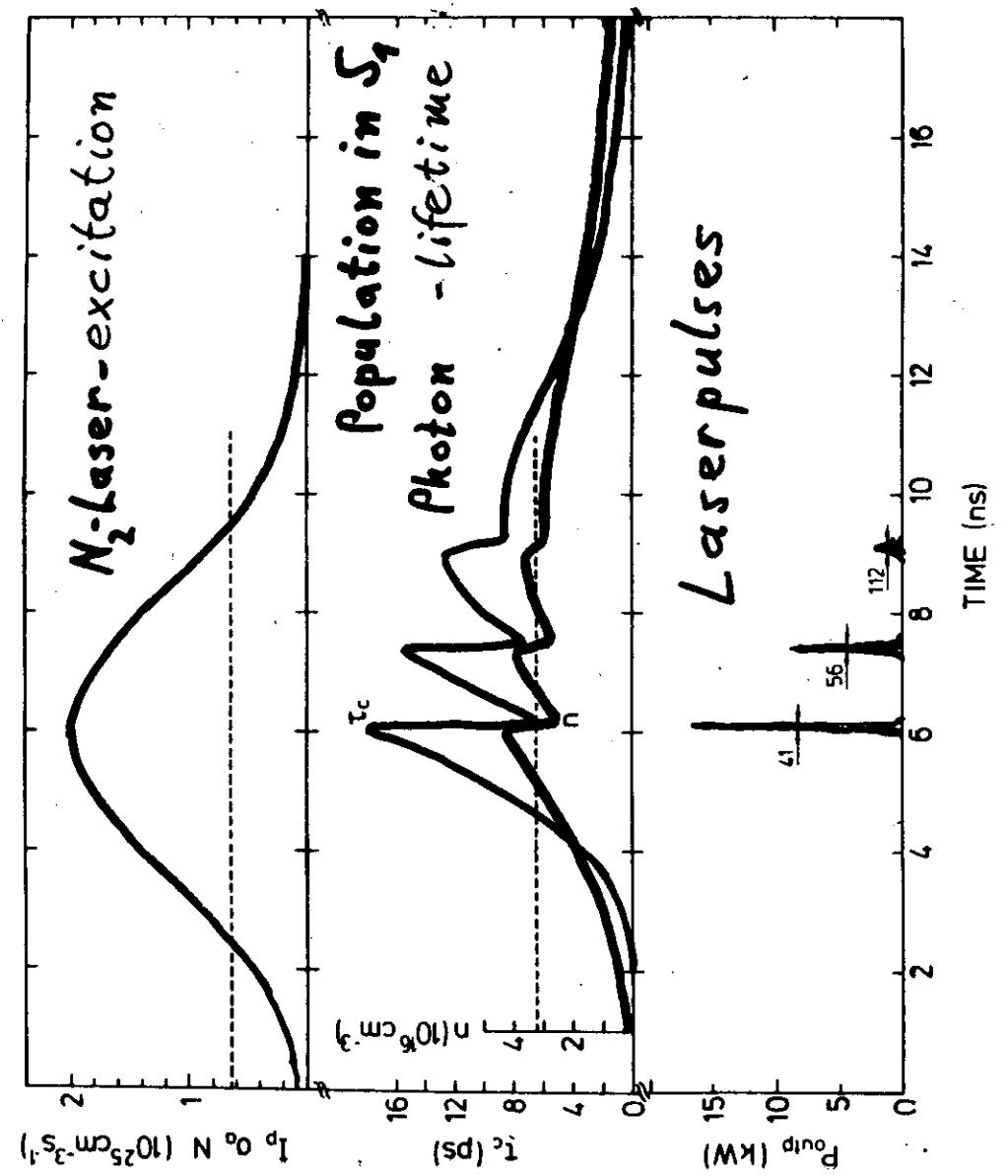




PRINCIPLE OF DISTRIBUTED FEEDBACK LASER

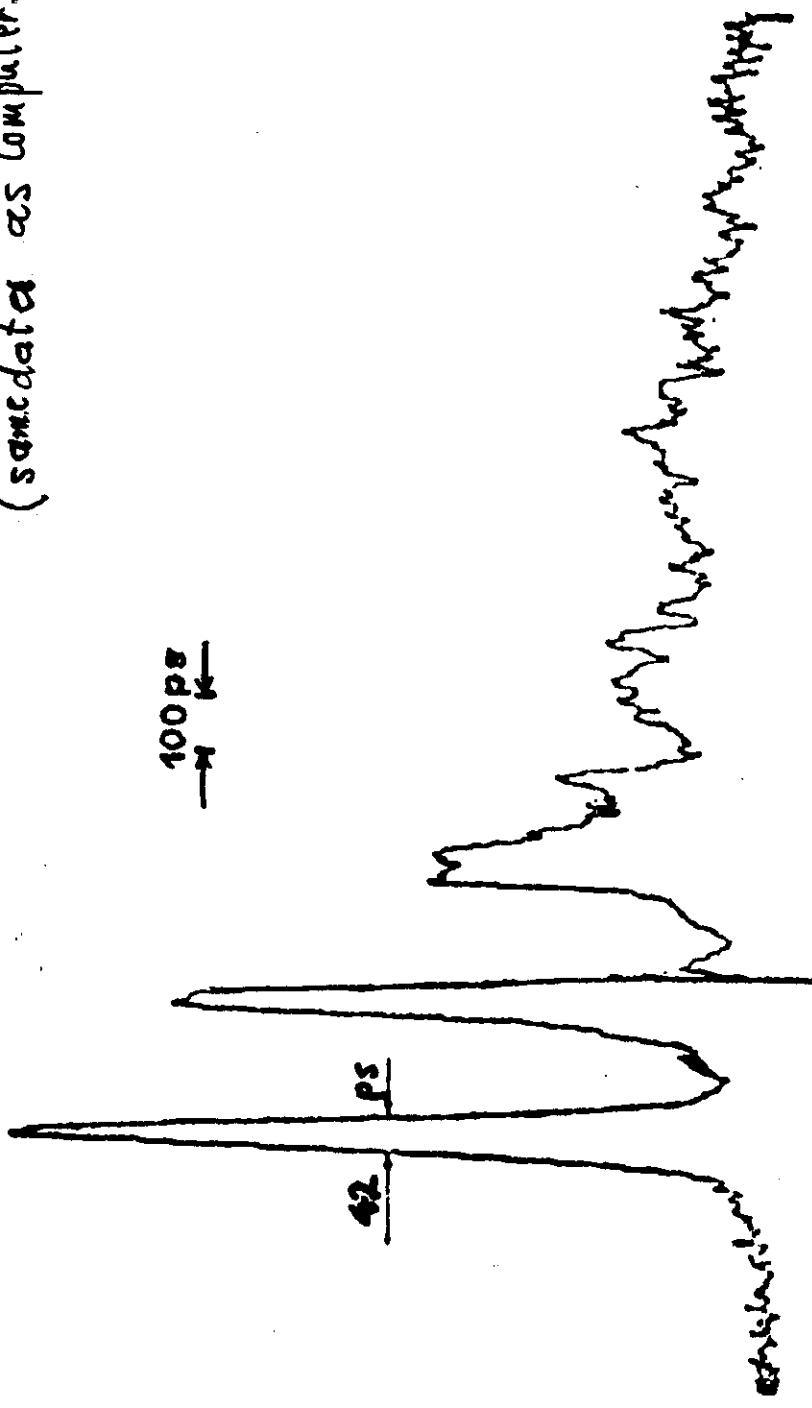
MEDIUM WITH SPATIALLY PERIODIC MODULATION OF
REFRACTIVE INDEX AND GAIN

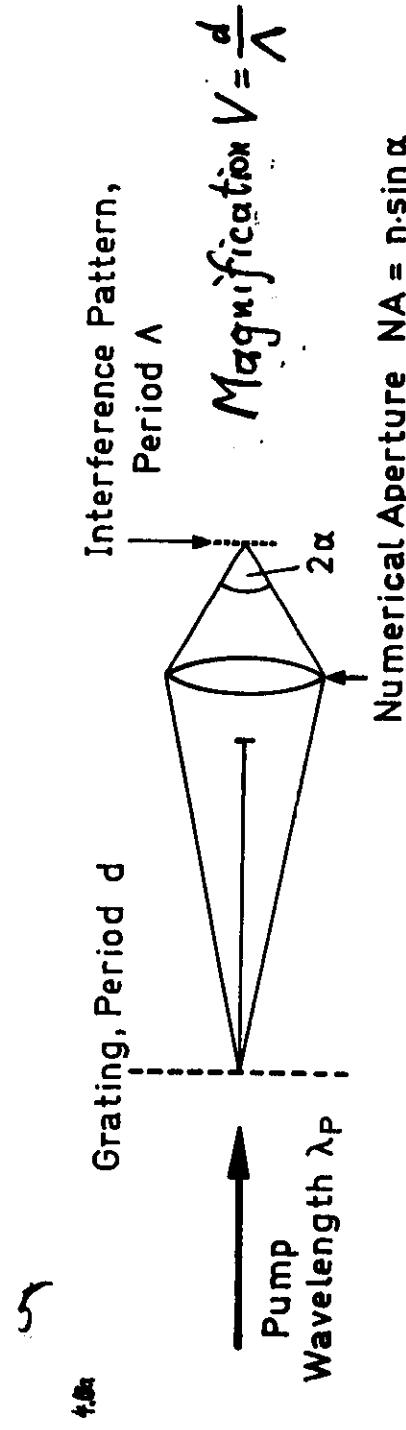
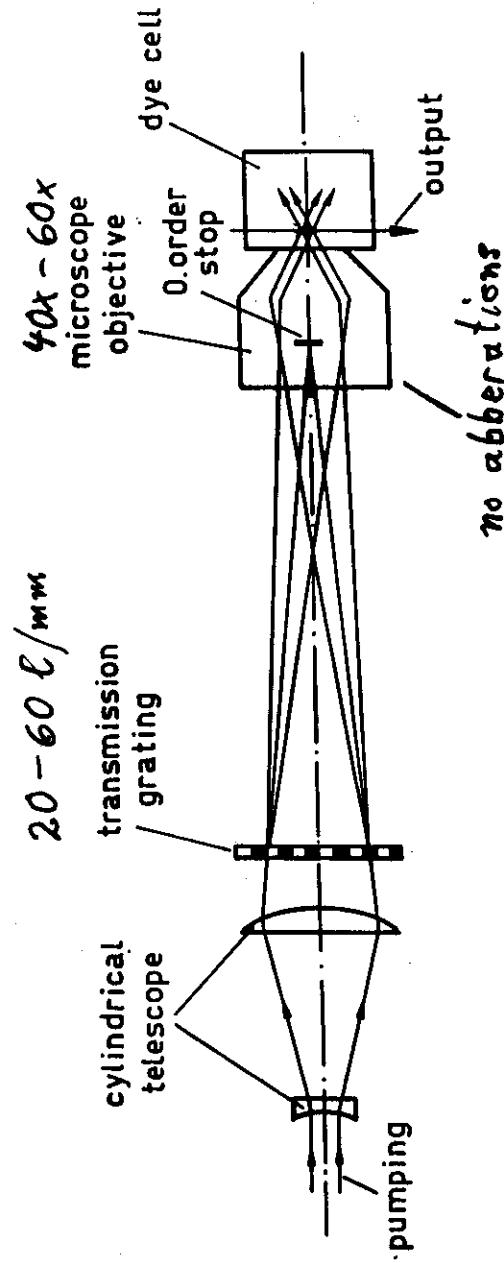




4.9

Densitometer curve of a streak-camera-record
(scanned as computer-solution)



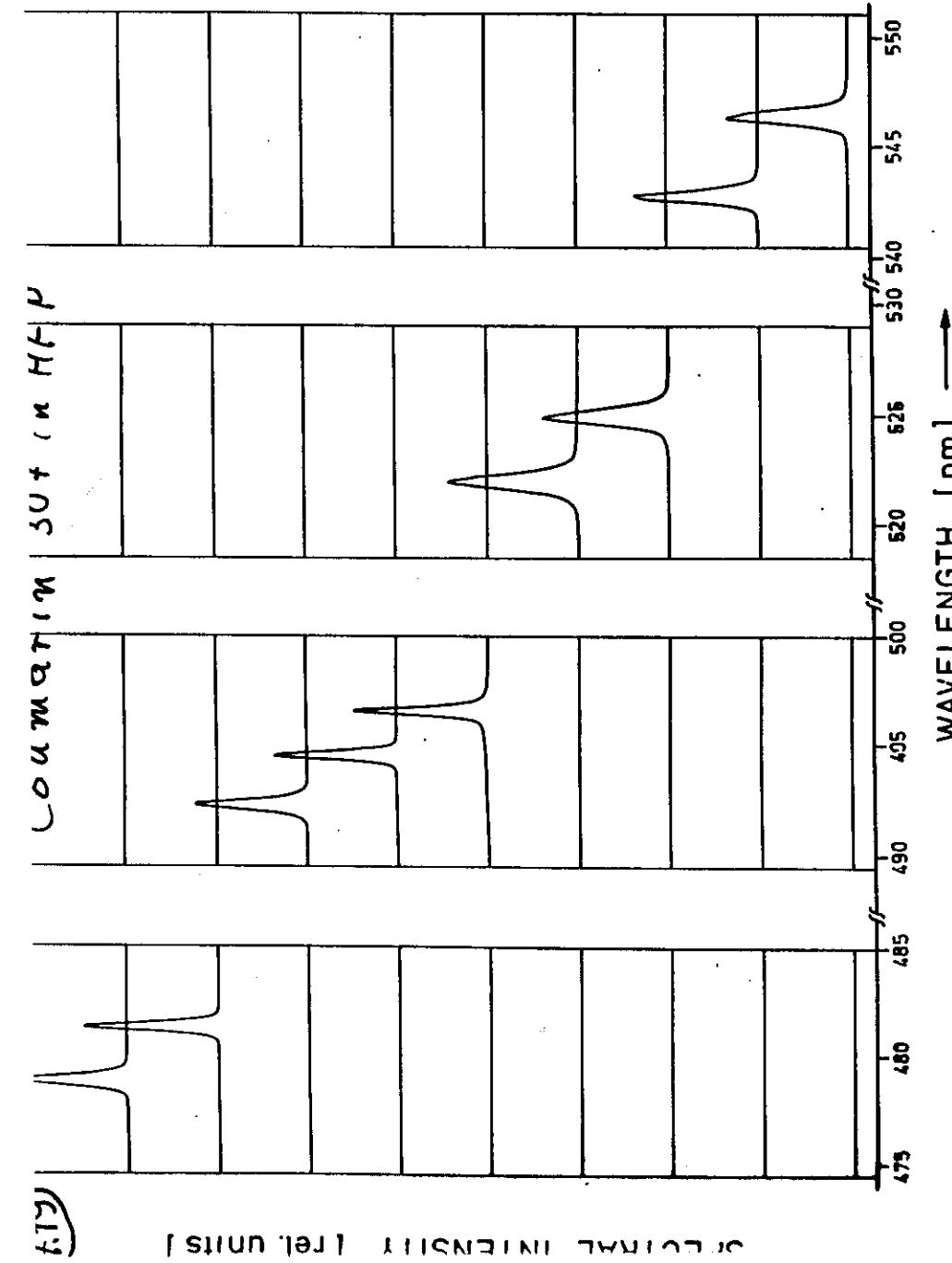


DFDL Wavelength $\lambda = 2n\lambda$; shortest wavelength: λ_{\min}

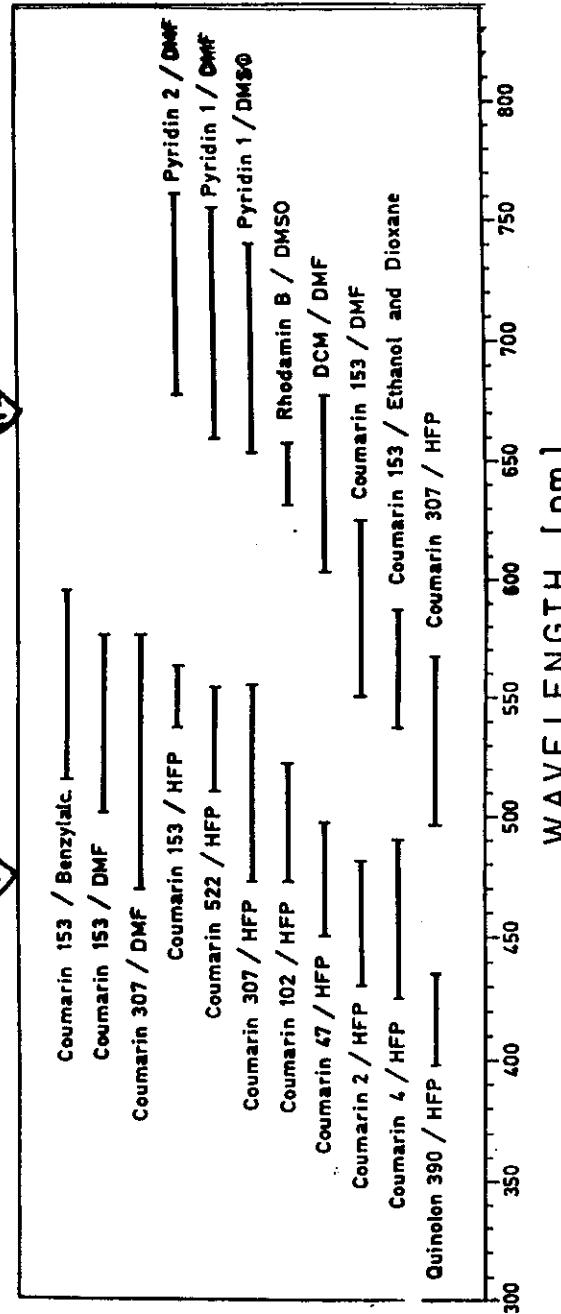
$$NA = n \cdot \lambda_p / \lambda_{\min}$$

Selection of microlens objective

Example:
 $\lambda_p = 360 \text{ nm}$
 $\lambda_{\min} = 380 \text{ nm}$
 $\longrightarrow NA = 1.23$



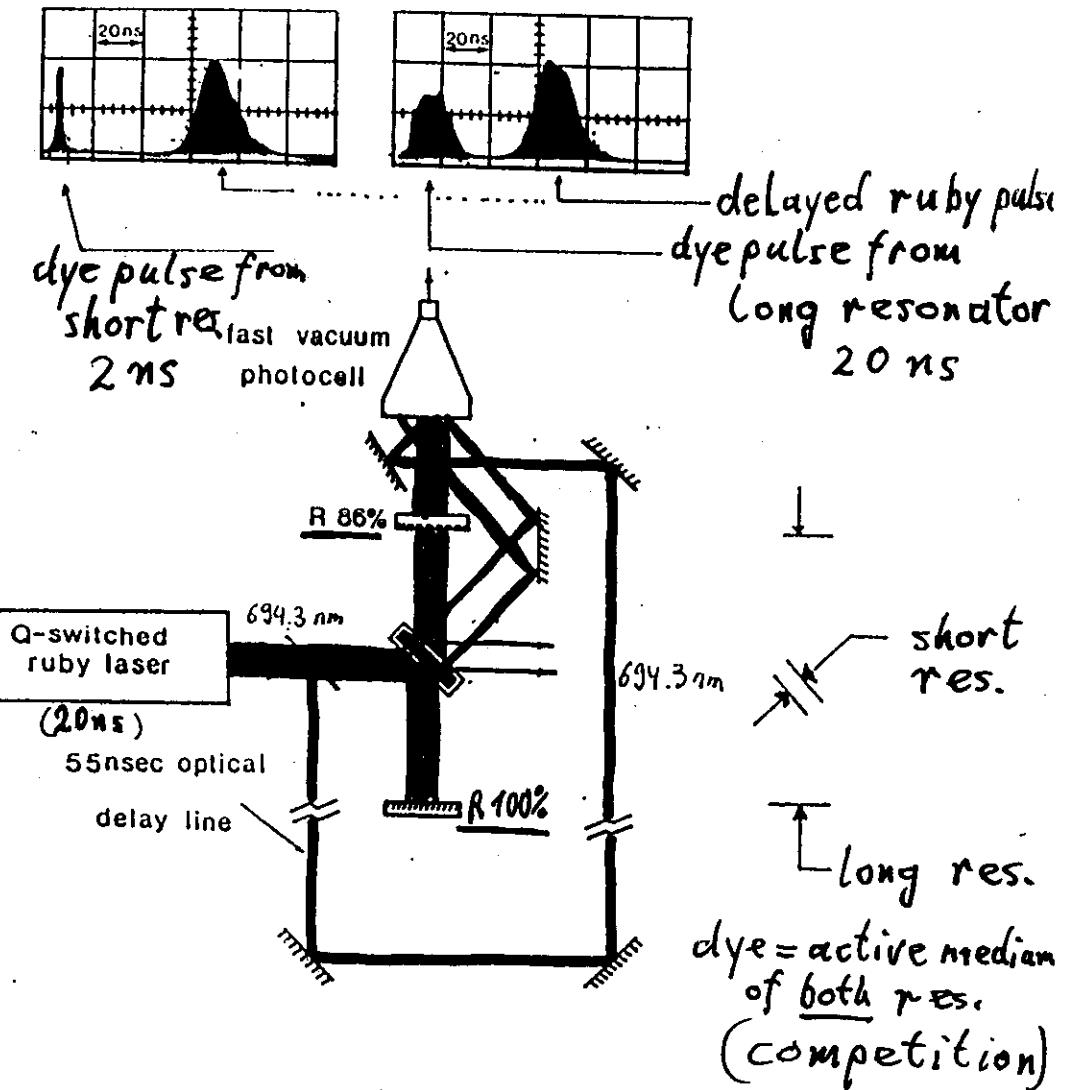
Zeiss 63x/1.2 Olympus 40x/1.00



Crossed Resonators

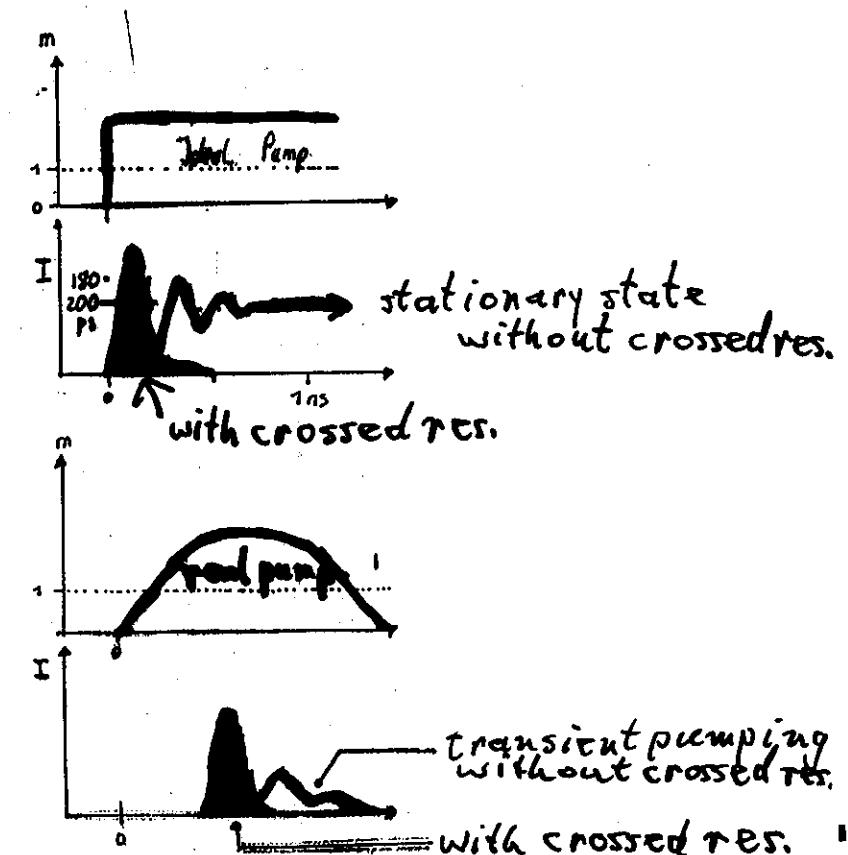
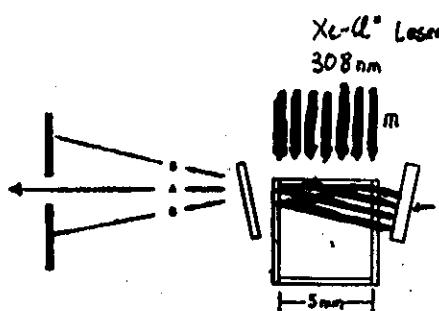
E. ERANIAN, P. DEZAUZIER, O. DE WITTE

OPT. COMMUN. 1 (1973) 150



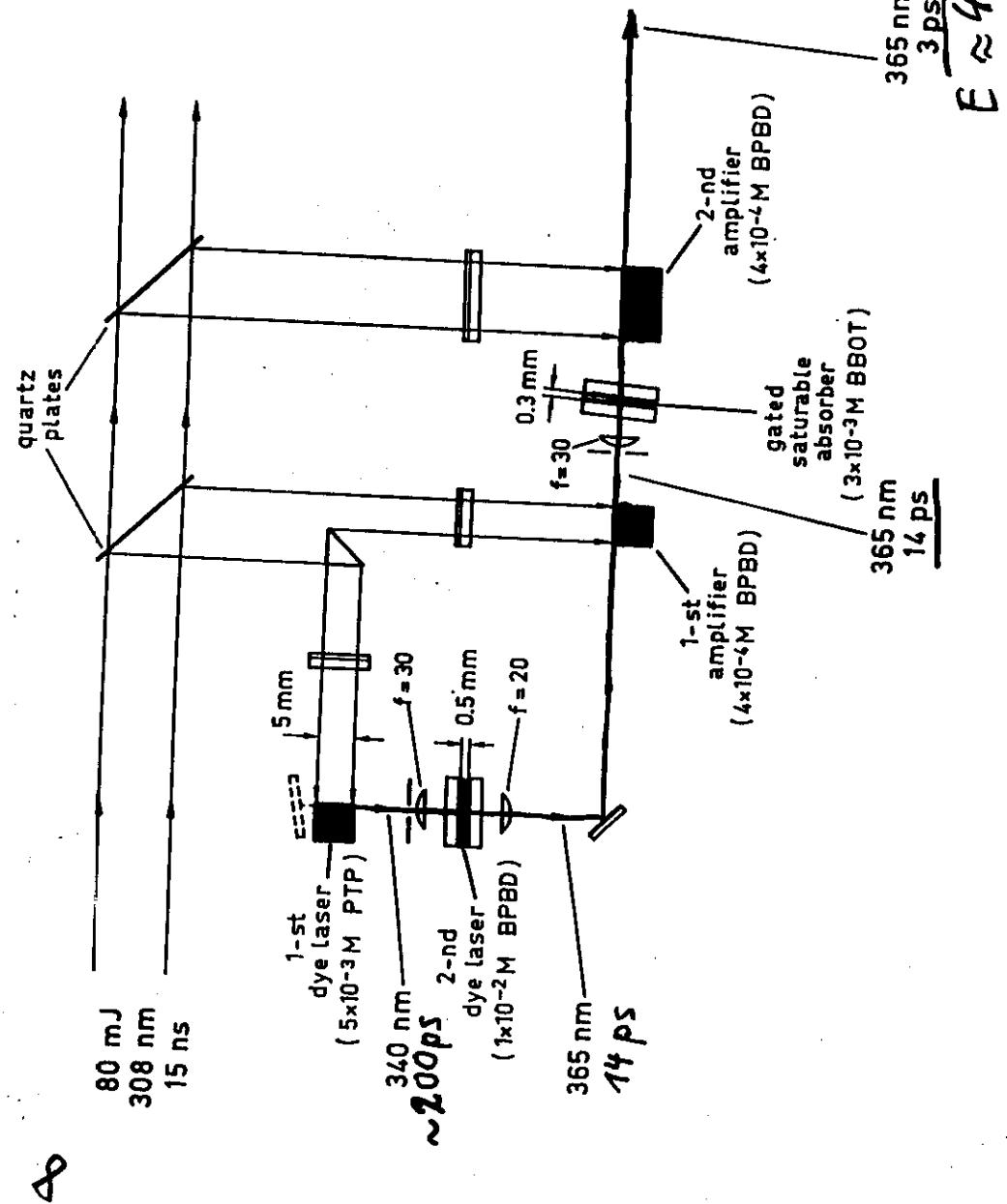
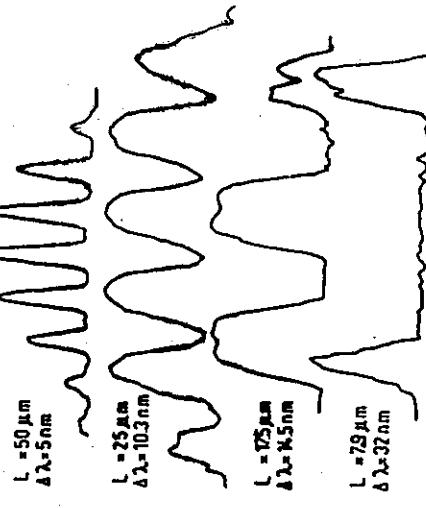
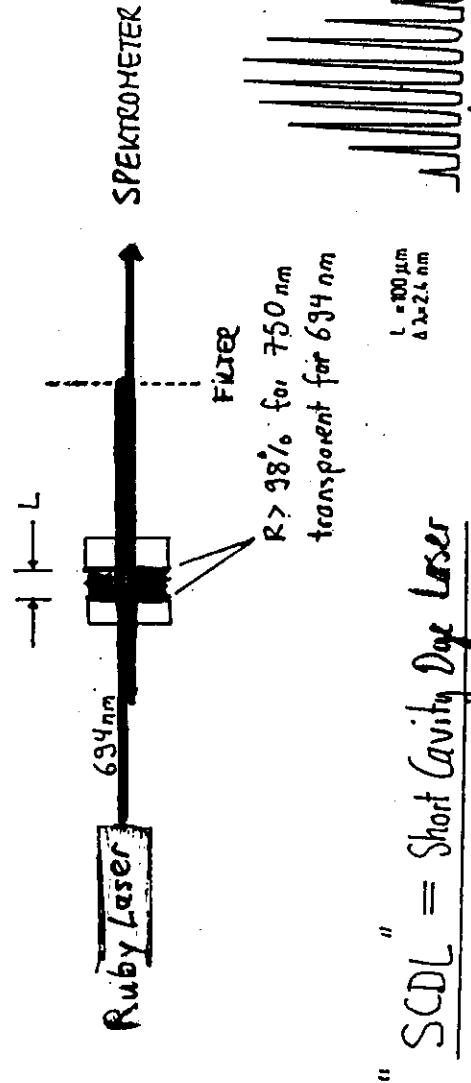
Transient pumping with crossed resonators

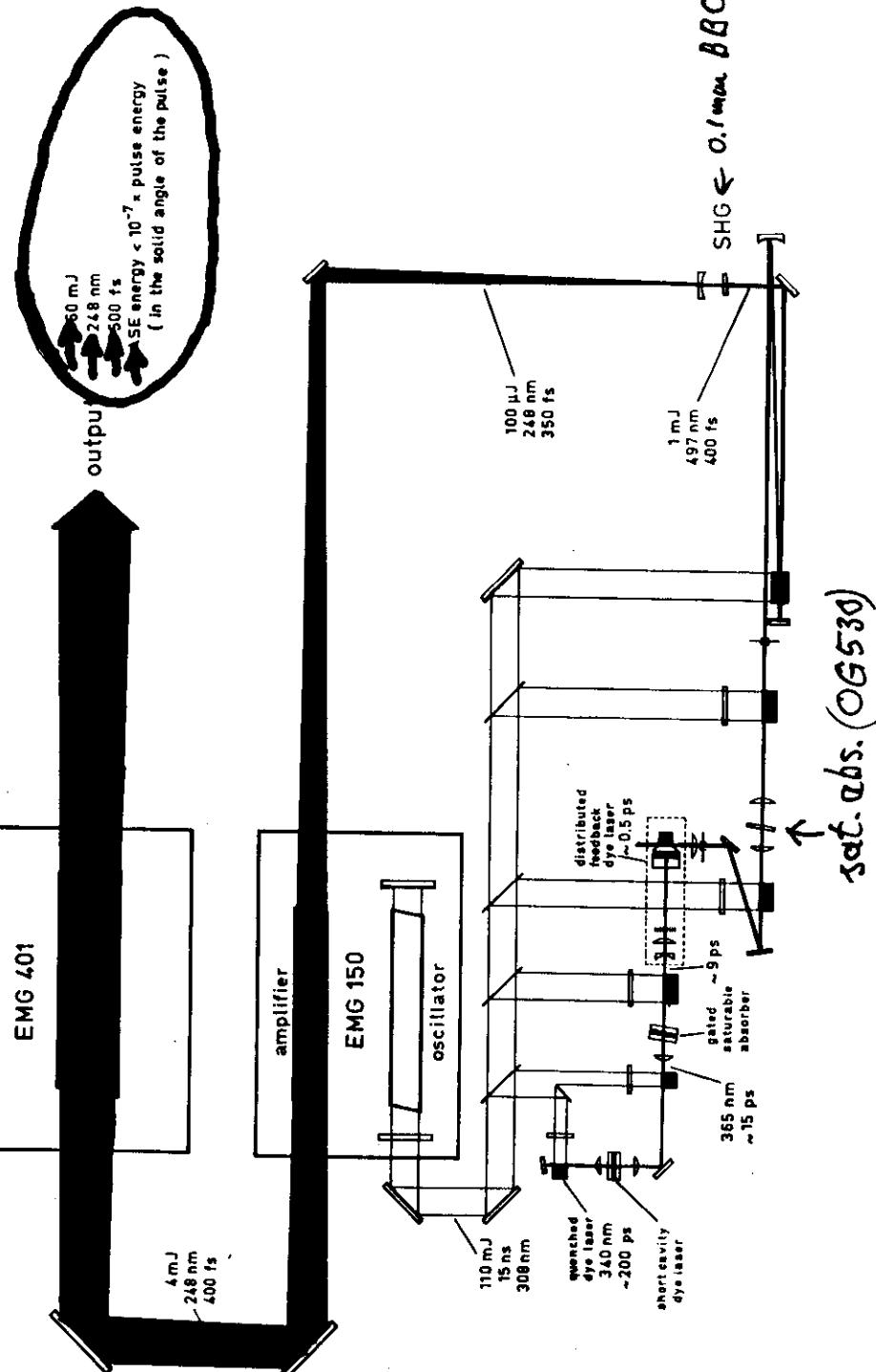
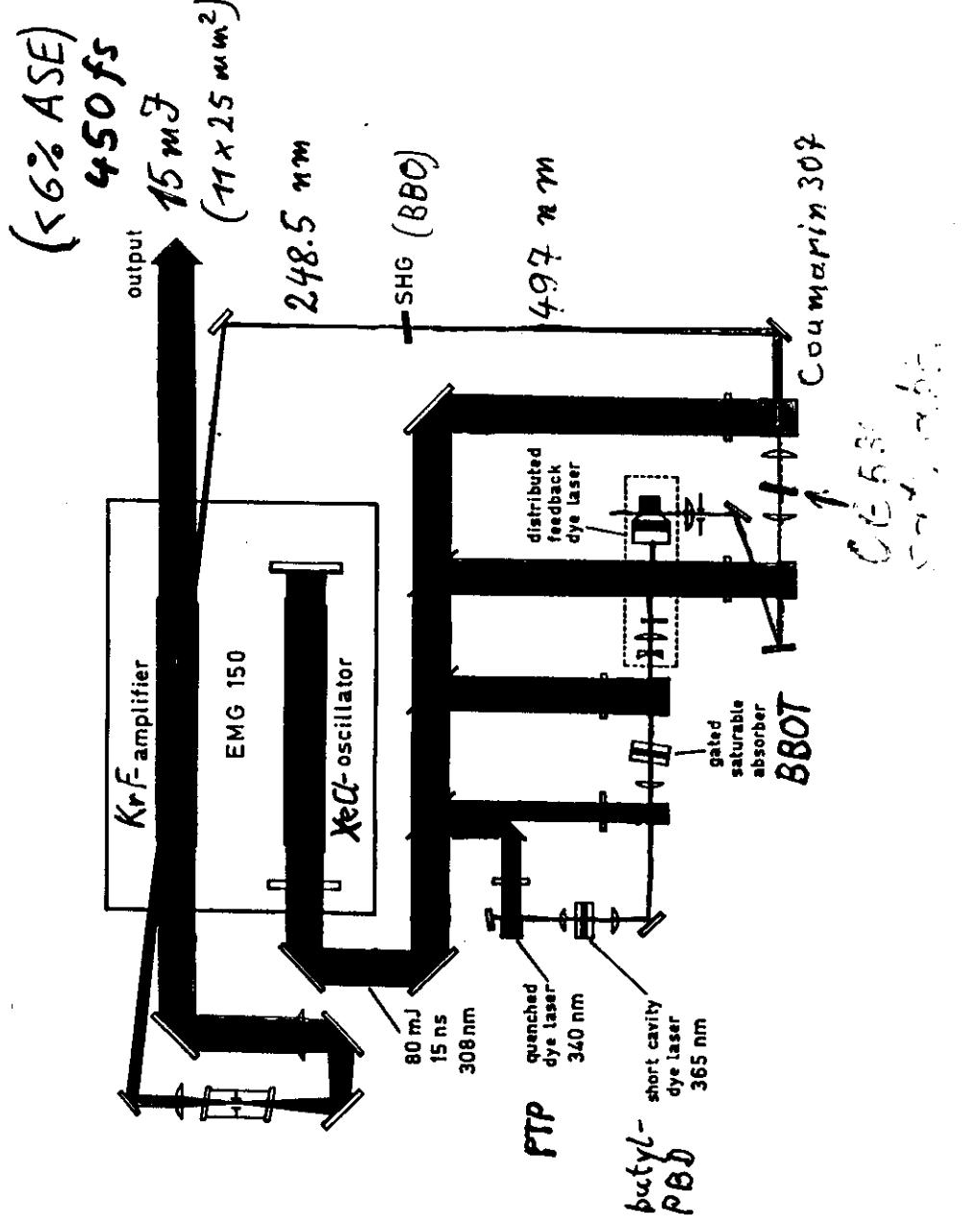
S. SETHMARÍ, F.P. SCHÄFER,
OPT. COMMUN. 28 (1983) 279

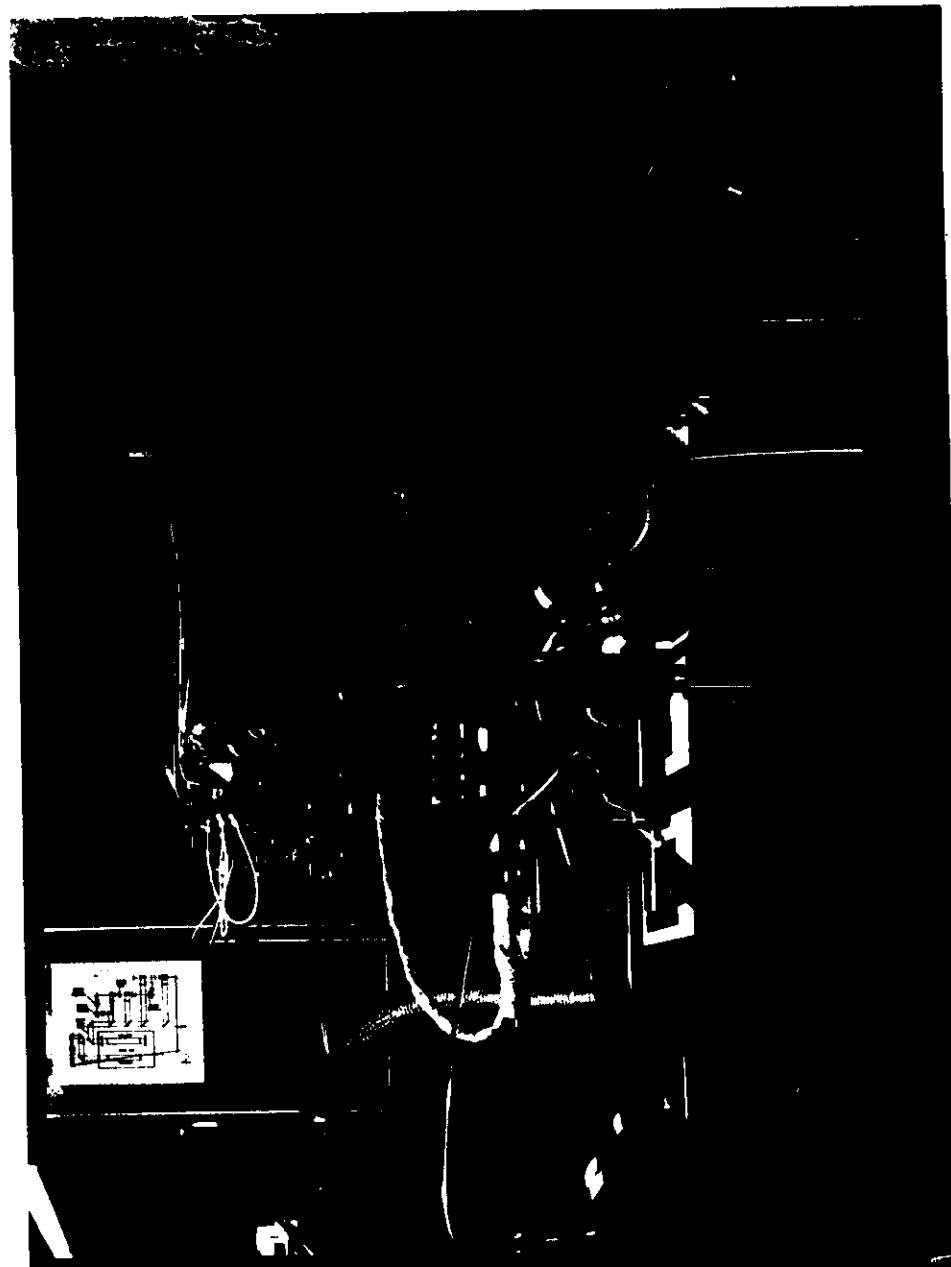
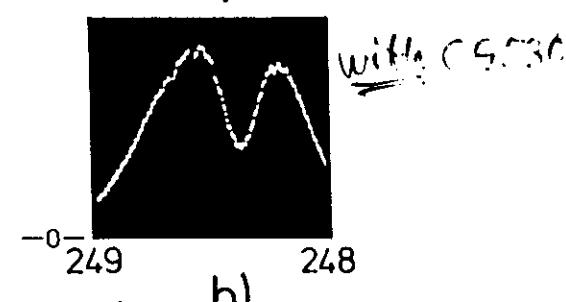
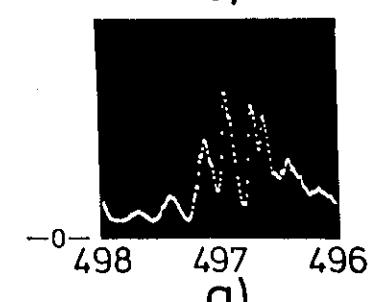
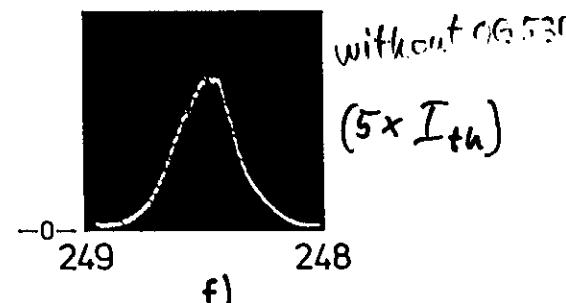
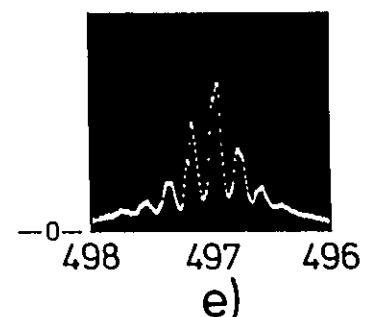
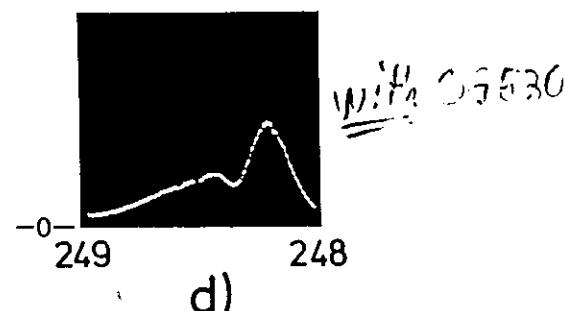
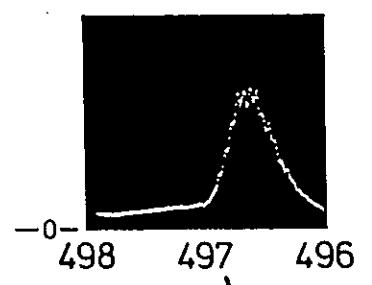
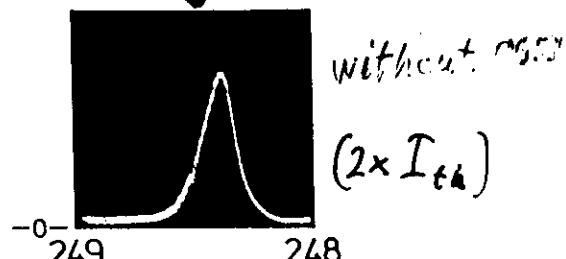
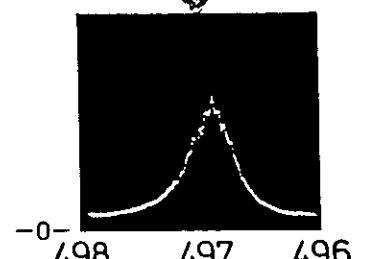


Wavelength-tuning by cavity length

F.P. SCHAFER, ANGEW. CHEMIE 82 (1970) 25

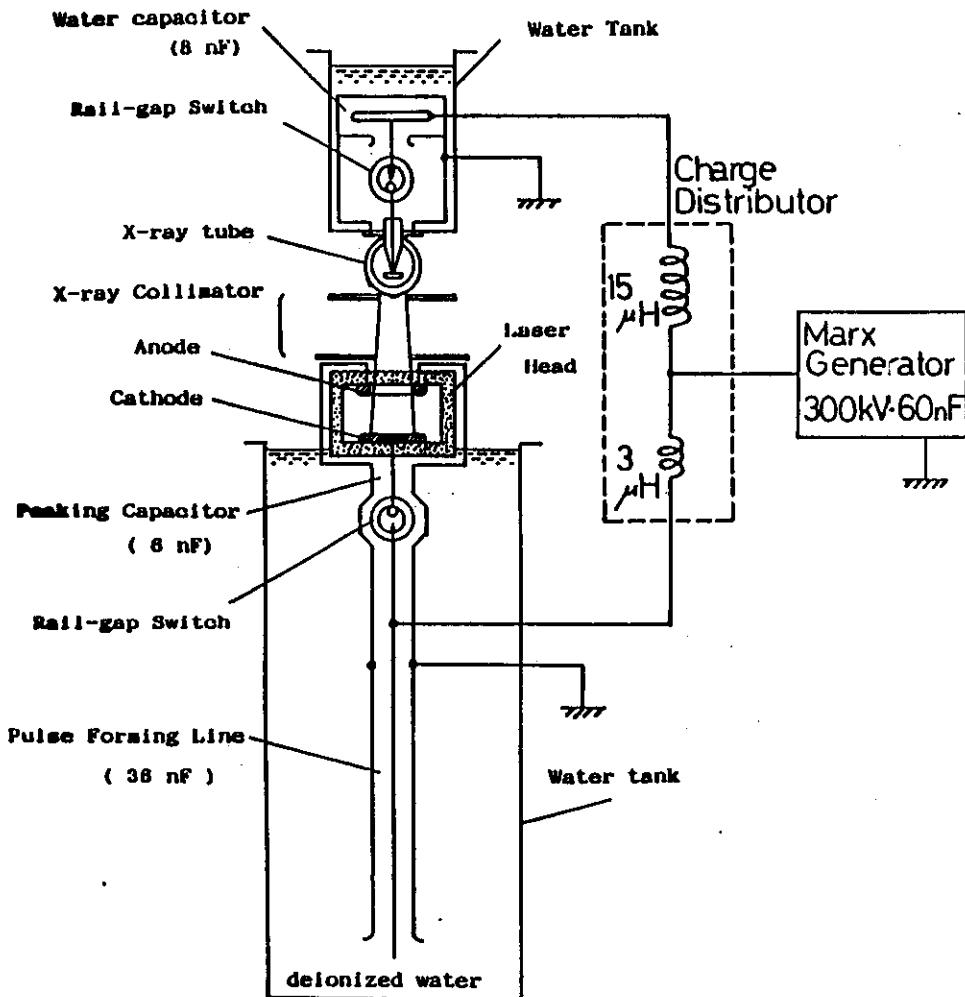




Spectra

KrF-Laser 10x8cm²

53 in 40ns, 1Hz



2nd Lecture:

- a) Measurement of Ultrashort UV Laser Pulses
- b) Handling of Ultrahigh Intensity Laser Pulses
- c) European Laser Facility SIMBA Project

Abstract. Pulsewidth measurements of ultrashort pulses in the ultraviolet need some modifications of the usual autocorrelation methods for the visible and infrared. The main point is replacing the second harmonic generation in crystals by multiple photon ionization in gases since no crystals are available for second harmonic generation in the region between 126 and 352 nm. For the shortest pulses of highest intensity also a special version of the usually applied Michelson interferometer is necessary to avoid 2-photon absorption in the beam splitter.

To reach shortest pulse duration it is often necessary to compress frequency chirped pulses after amplification in the ultraviolet and at high intensities. The best method for this is the use of a prism compressor made up of two prisms of a suitable material like LiF and a retro-reflector.

When applying very short pulses one has to take into account the group velocity dispersion in optical materials which is especially pronounced in the ultraviolet. The dramatic effects of pulse front distortion by group velocity dispersion will be demonstrated together with methods of compensating these effects. Another very important effect at highest intensities in the ultraviolet is multiple photon absorption in optical materials which we measured exactly. For thick samples of optical materials other nonlinear effects also play an important role like self-phase modulation and self-focussing which will be briefly discussed.

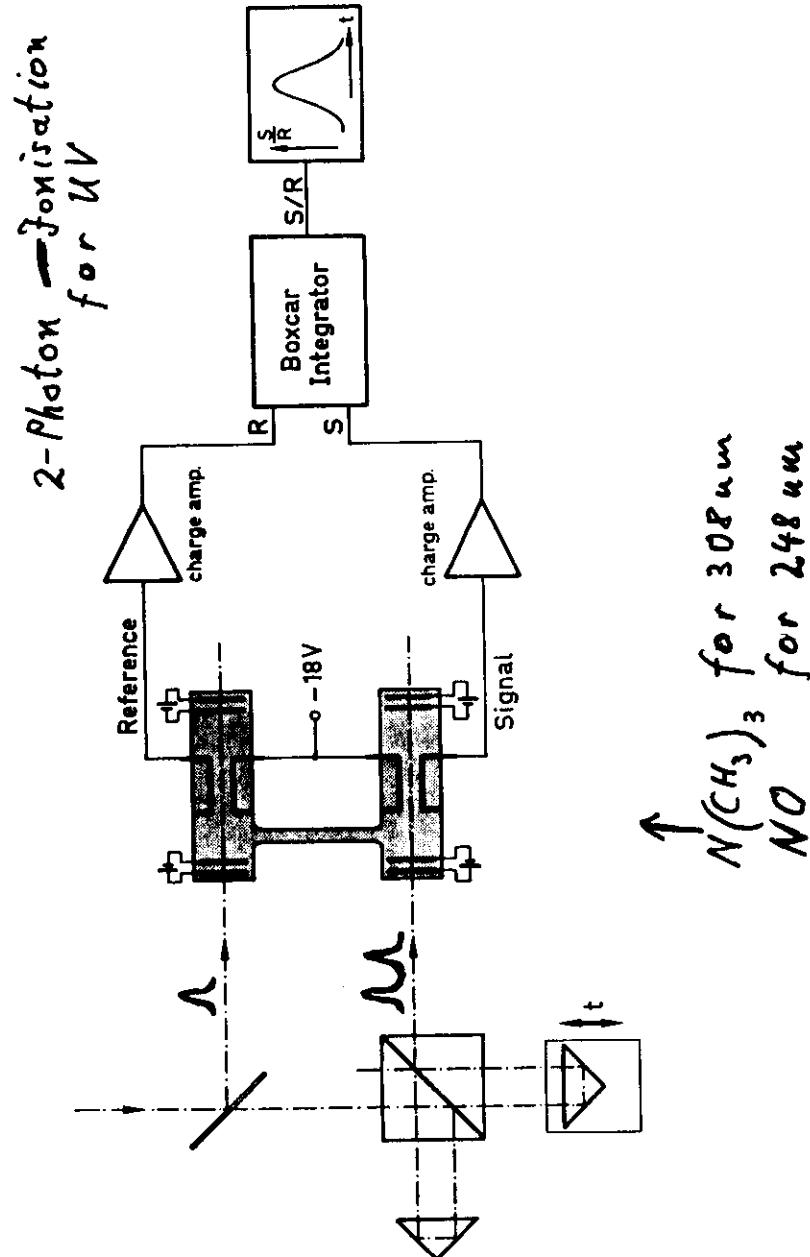
We recently proposed to build a 100 J/100 fs KrF-laser in the frame work of a European High Performance Laser Facility. The constructional principles of this laser and the technical difficulties connected with its realization will be briefly discussed. Some examples of new experiments that will become possible with this laser will be given.

2nd Lecture

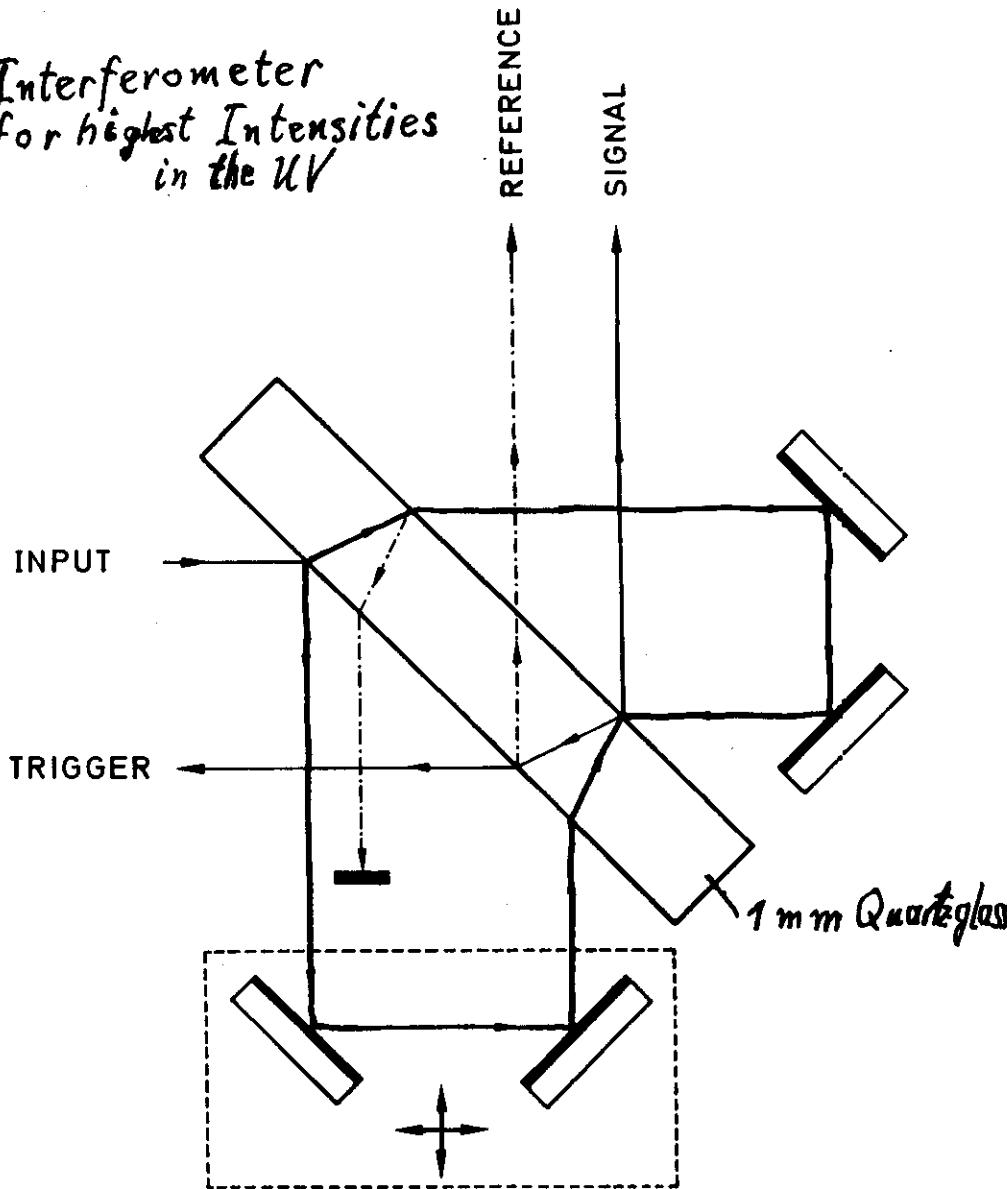
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window.
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Pulse front and pulse duration distortion in refractive optics, and its
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- 2.5 P. Simon, H. Gerhardt, S. Szatmári:
Intensity-dependent loss properties of window materials at 248 nm.
Opt. Lett. 14, 1207-1209 (1989)

Sampling - method



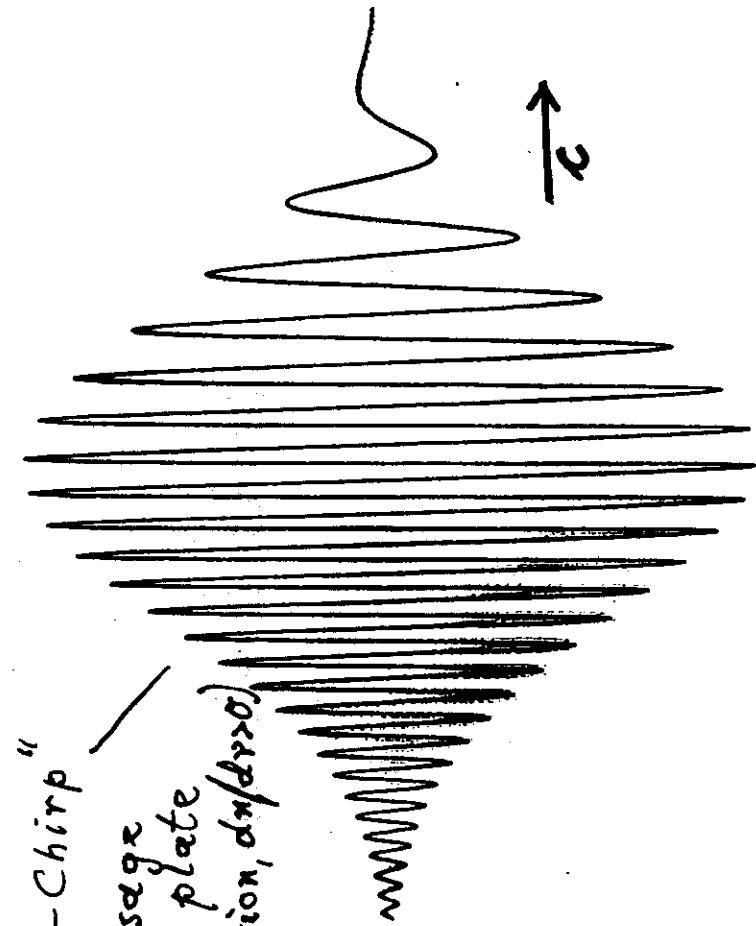
Interferometer
for highest Intensities
in the UV



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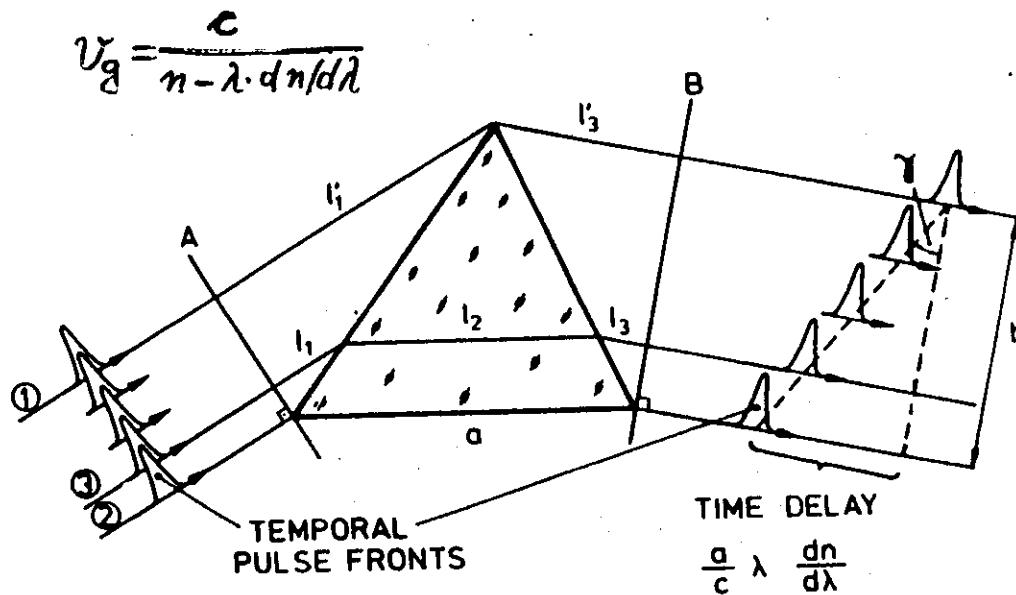
Frequency-modulated Gaussian pulse

$$\Delta t_{FWHM} \times 4\nu_{FWHM} > 0.441$$

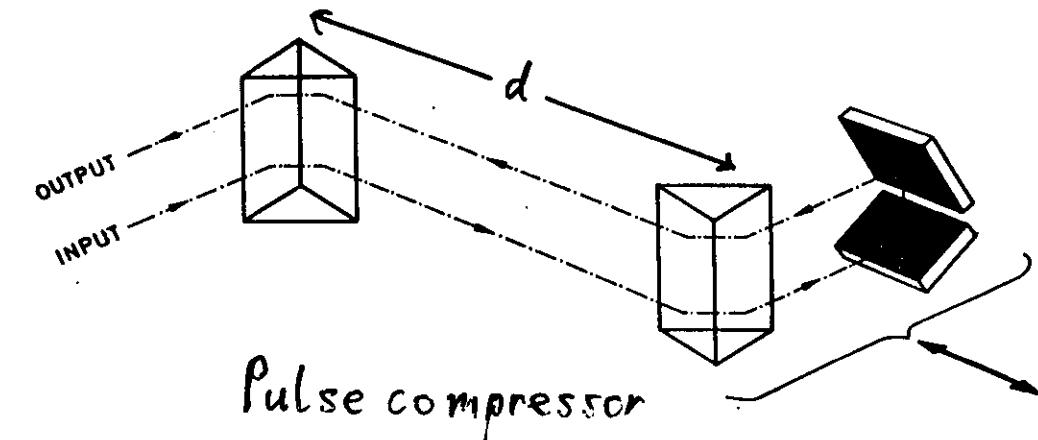
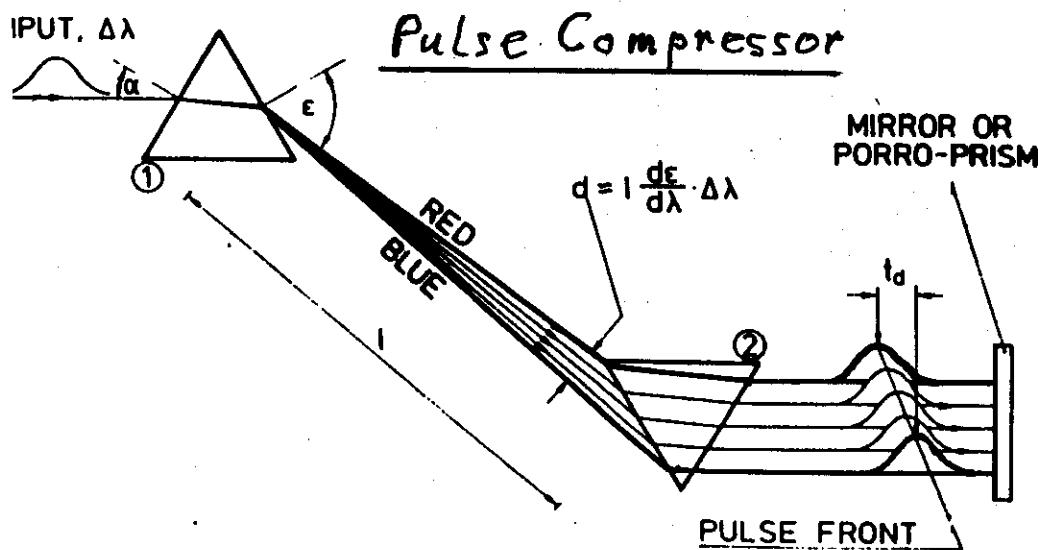
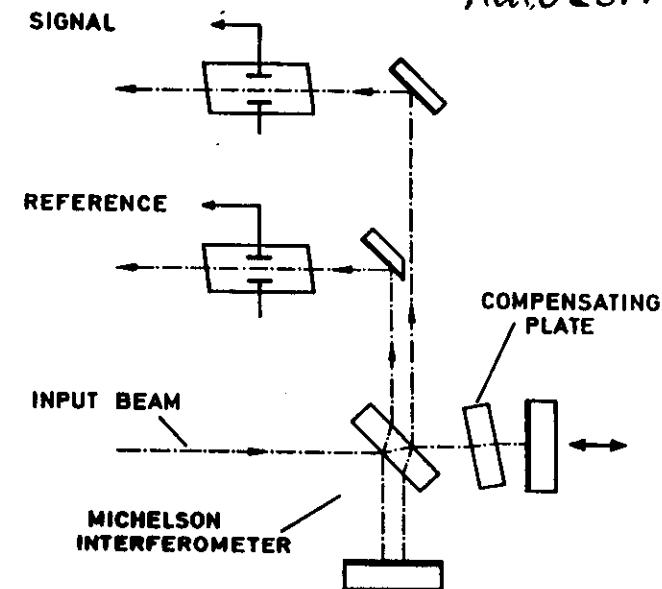


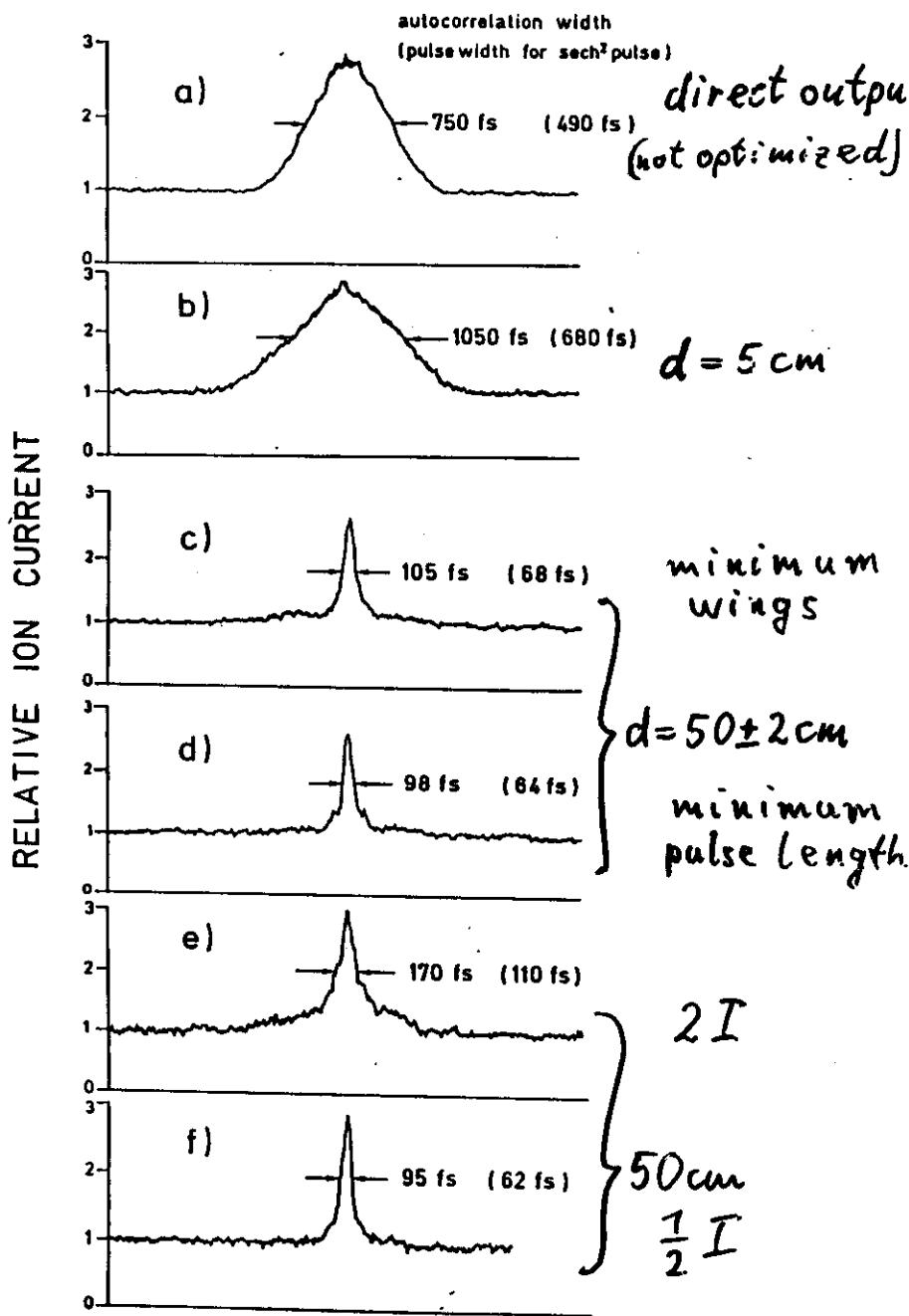
Group velocity v_g

5.28 6.8



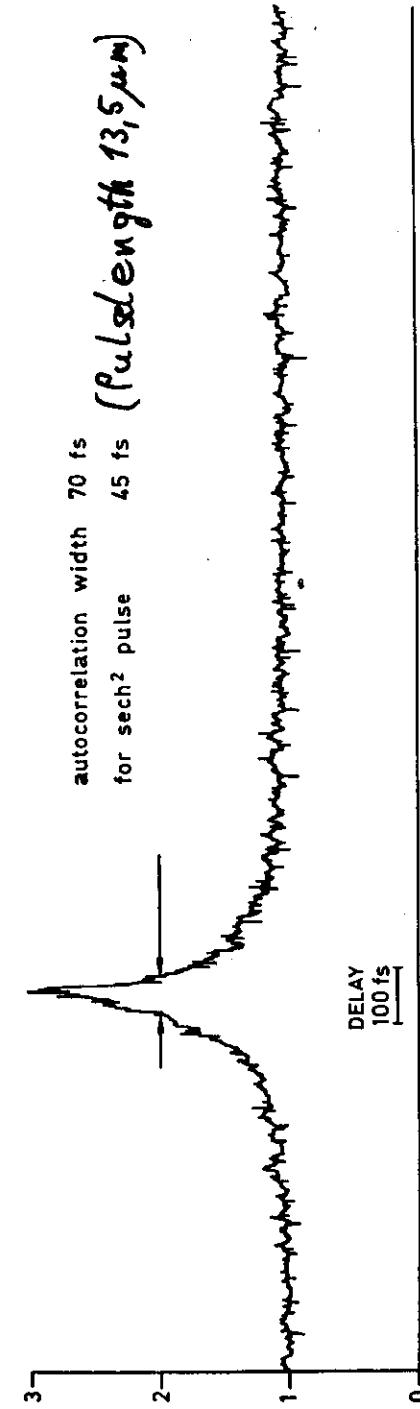
Autocorrelator





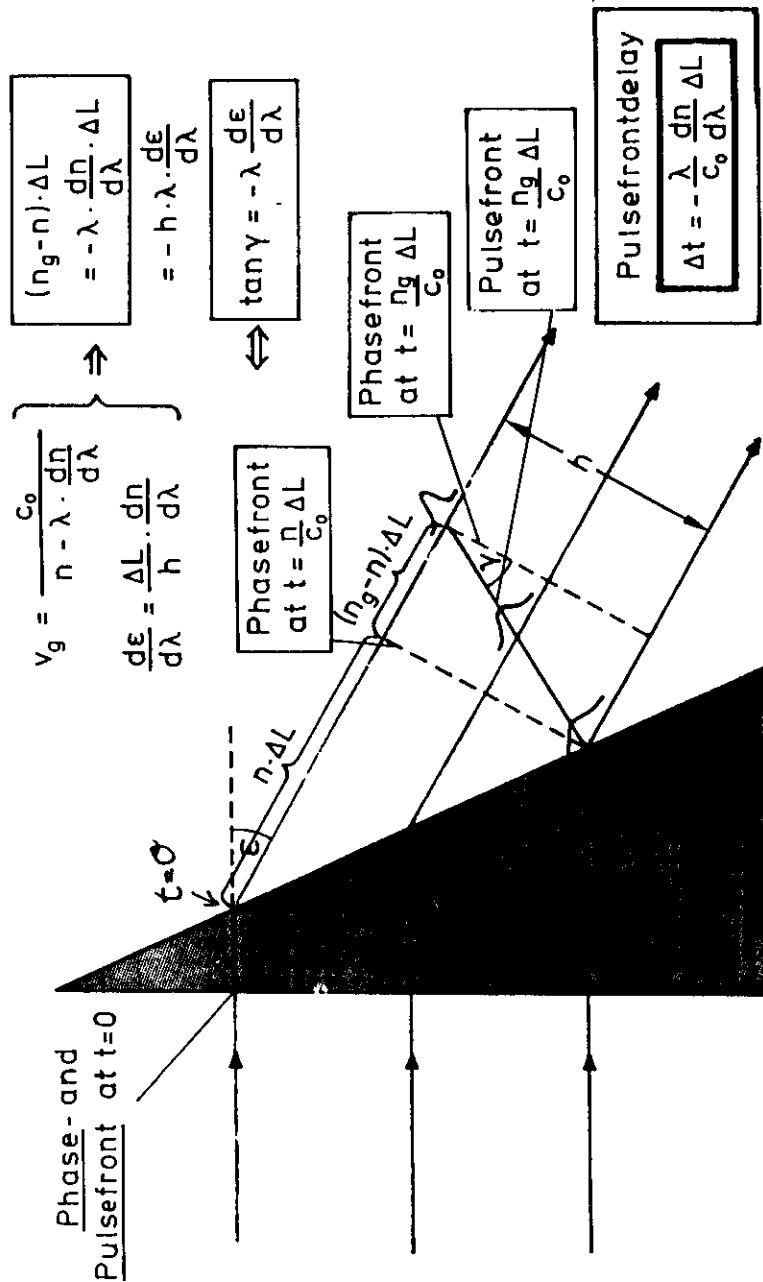
Szatmari, Schäfer: Simplified
Laser System for the Generation ...
Fig. 5

75



Pulse power $> 1 \text{ TW}$

Pulse front vs. Phase front

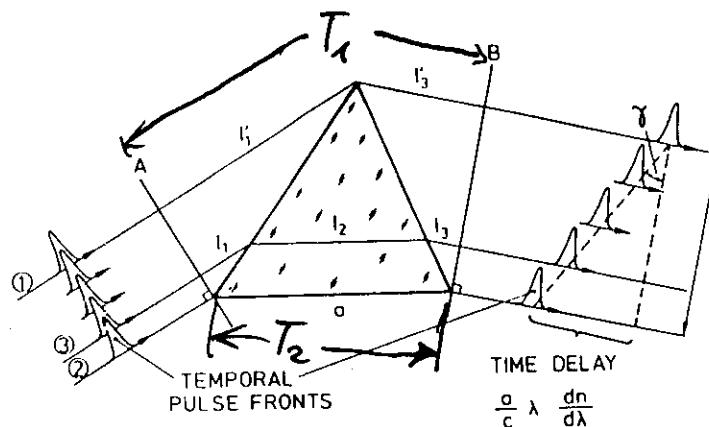


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Group velocity v_g

$$v_g = c / (n - \lambda dn/d\lambda)$$

$$\left. \begin{array}{l} T_1 = l_1'/c + l_3'/c = a \cdot n/c \\ T_2 = \frac{a}{c(n - \lambda dn/d\lambda)} \end{array} \right\} \Delta T = \frac{a}{c} \lambda \frac{dn}{d\lambda}$$



Numerical example:

Suprasil: $\lambda = 249 \mu m$, $n = 1,508$ $\frac{dn}{d\lambda} = 5,46 \cdot 10^{-4} \mu m^{-1}$

$$a = 5 \text{ cm} \Rightarrow \Delta T = 22,7 \text{ ps} \approx 6,8 \text{ mm}$$

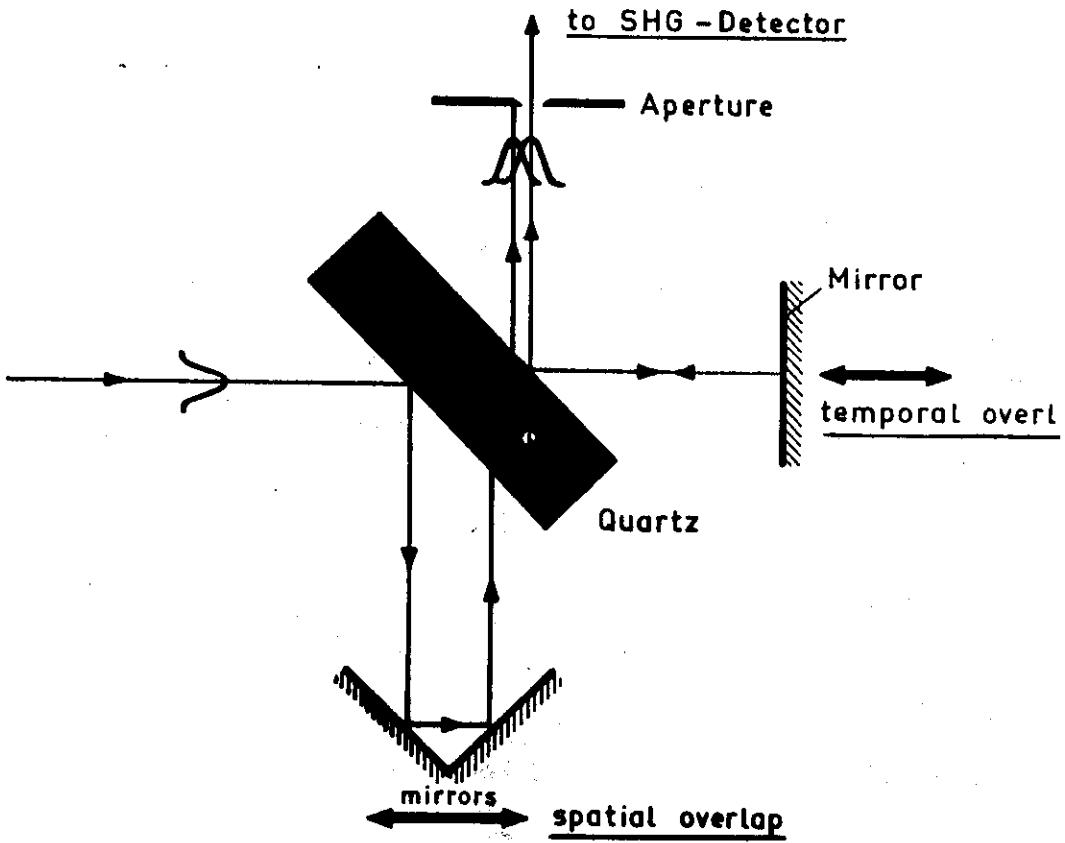
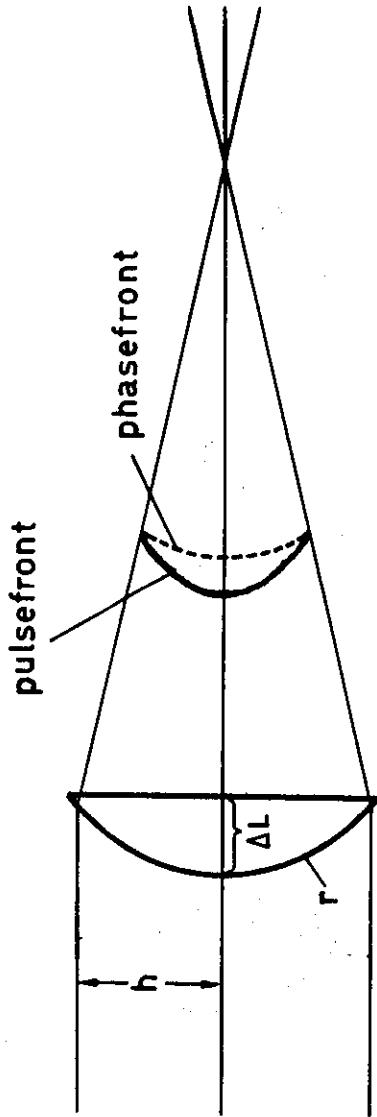
$$v_p = \frac{c}{n} = 1,99 \cdot 10^{10} \frac{\text{cm}}{\text{s}}$$

$$v_g = \frac{c}{n - \lambda dn/d\lambda} = 1,82 \cdot 10^{10} \frac{\text{cm}}{\text{s}}$$

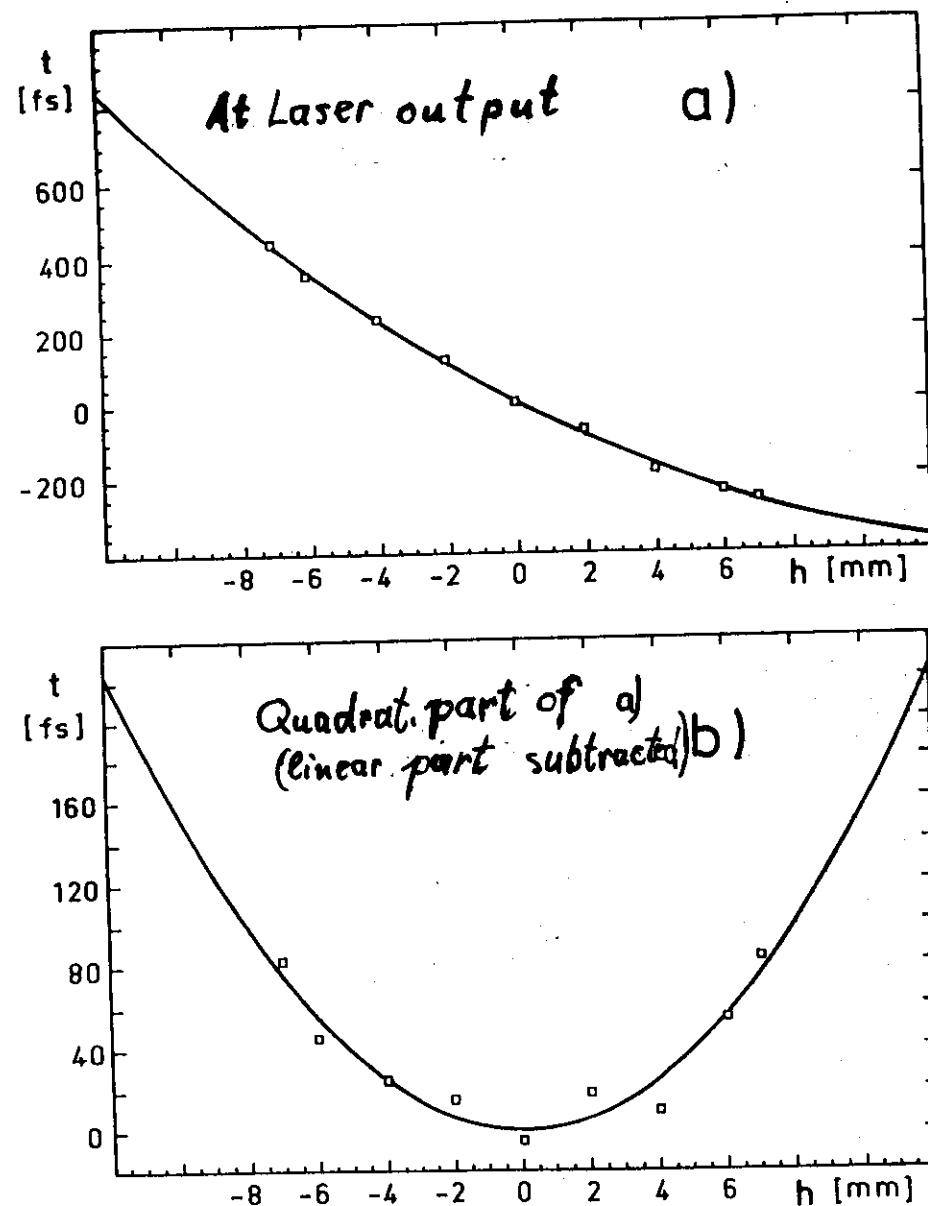
$$\frac{v_p}{v_g} = 1,09$$

Autocorrelator for Measurement of Pulse-Front Curvature

Pulse-front distortion
by dispersion in focussing lenses

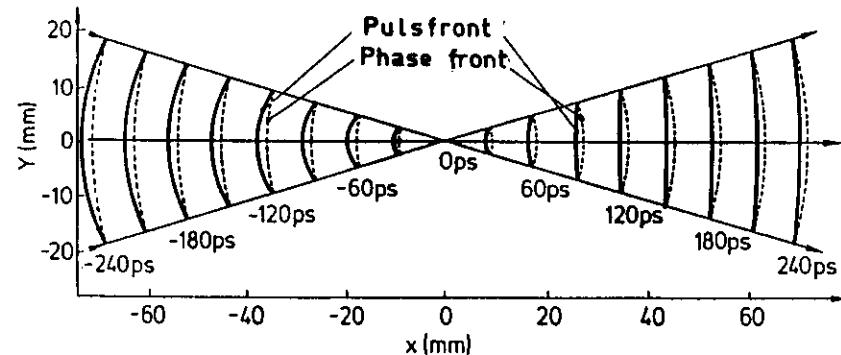


(S. Szatmári, G. Kühnle, Opt. Commun. 69 (1988) 60

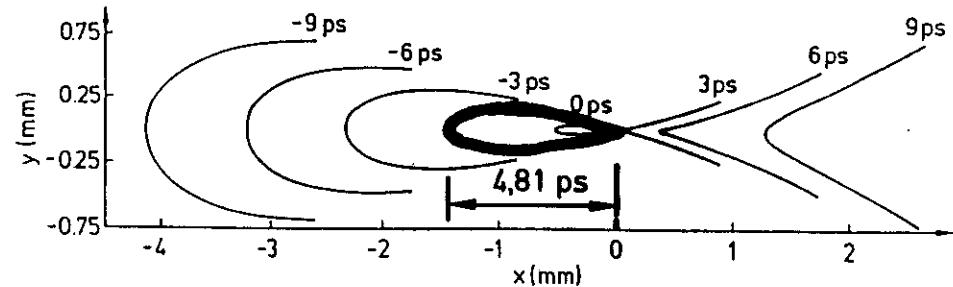


Apparent pulse lengthening on target
($f=50\text{ mm}$, $R=40\text{ mm}$, $\lambda=248\text{ nm}$; quartz glass)

a)

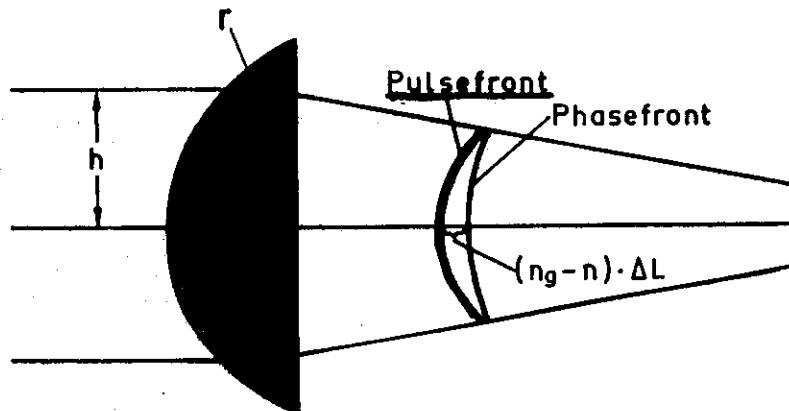


b)



(Zs. Bor, J. modern optics 35(1988) 1907)

Thin Lenses and Telescopes



$$\text{IF: } \left(\frac{h}{r}\right)^2 \ll 1 :$$

$$\Delta L \approx r \cdot \left(1 - \sqrt{1 - \left(\frac{h}{r}\right)^2}\right) \approx \frac{h^2}{2r}$$

Kepler - Telescope :

$$\Delta L \approx \frac{h_{in}^2}{2r_{in}} (1 + M)$$

Galilei - Telescope :

$$\Delta L \approx \frac{h_{in}^2}{2r_{in}} (1 - M)$$

19

58)

Pulsefront - corrected Optics

Examples of corrected optics:

- Reflective Optics
- Lens systems made from different materials
(Achromats)
- Normal lenses and an additional compensation element

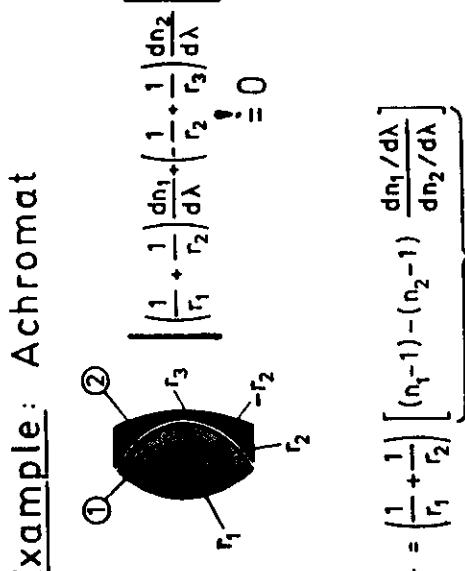
Lens system:

$$\Delta t = -\frac{\lambda}{c_0} \frac{h^2}{2} \frac{1}{r} \frac{dn}{d\lambda}$$

General formula for a thin lens system
with $\left(\frac{h_i}{r_i}\right)^2 \ll 1$: $\Delta t = -\frac{\lambda}{c_0} \frac{h^2}{2} \sum_i \frac{1}{r_i} \frac{dn_i}{d\lambda}$

⇒ Pulsefront - correction means :

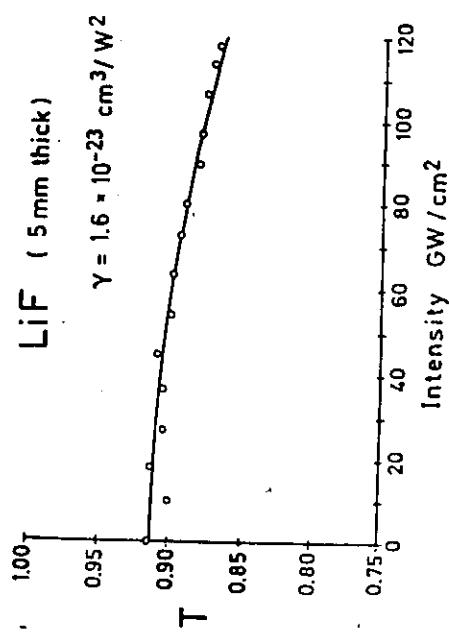
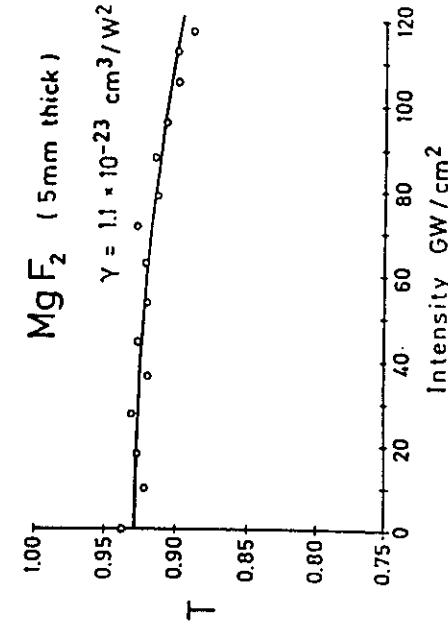
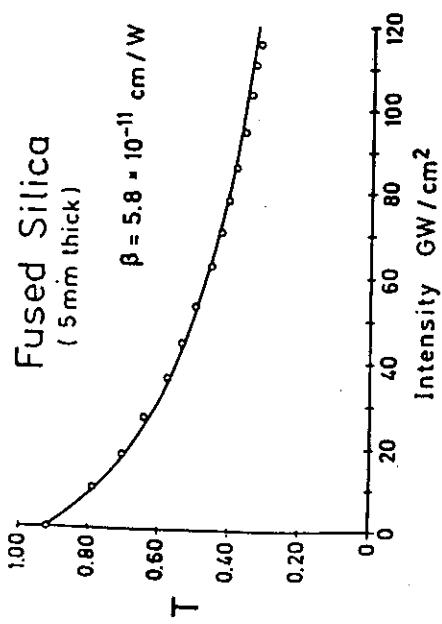
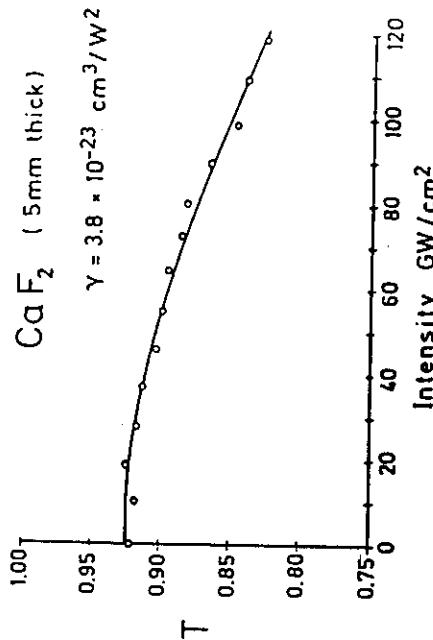
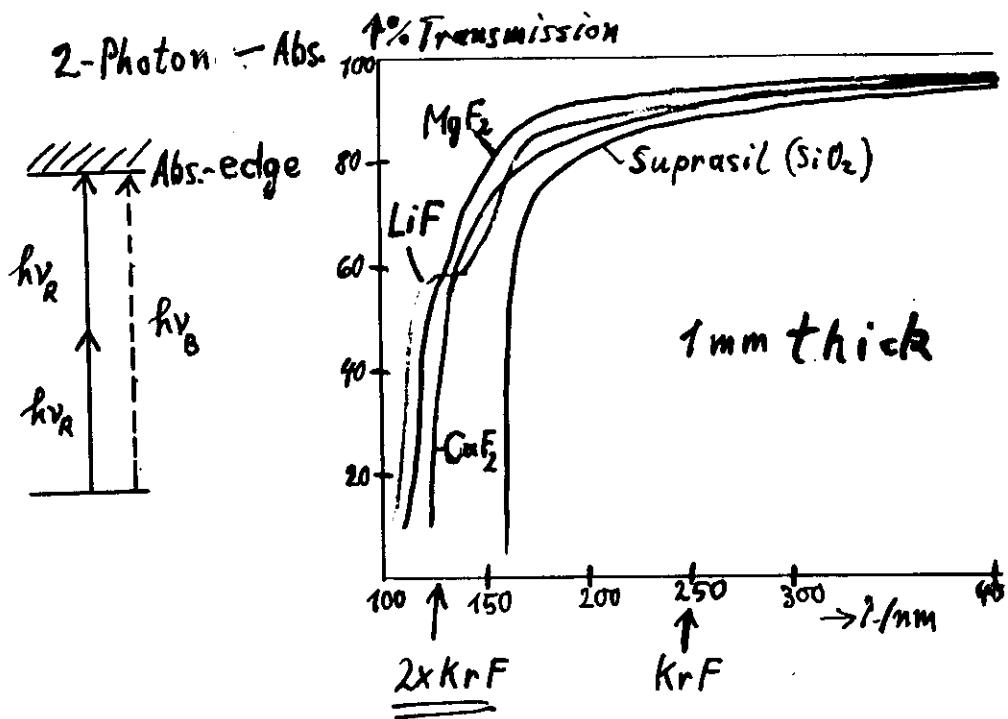
$$\sum_i \frac{1}{r_i} \cdot \frac{dn_i}{d\lambda} = 0$$



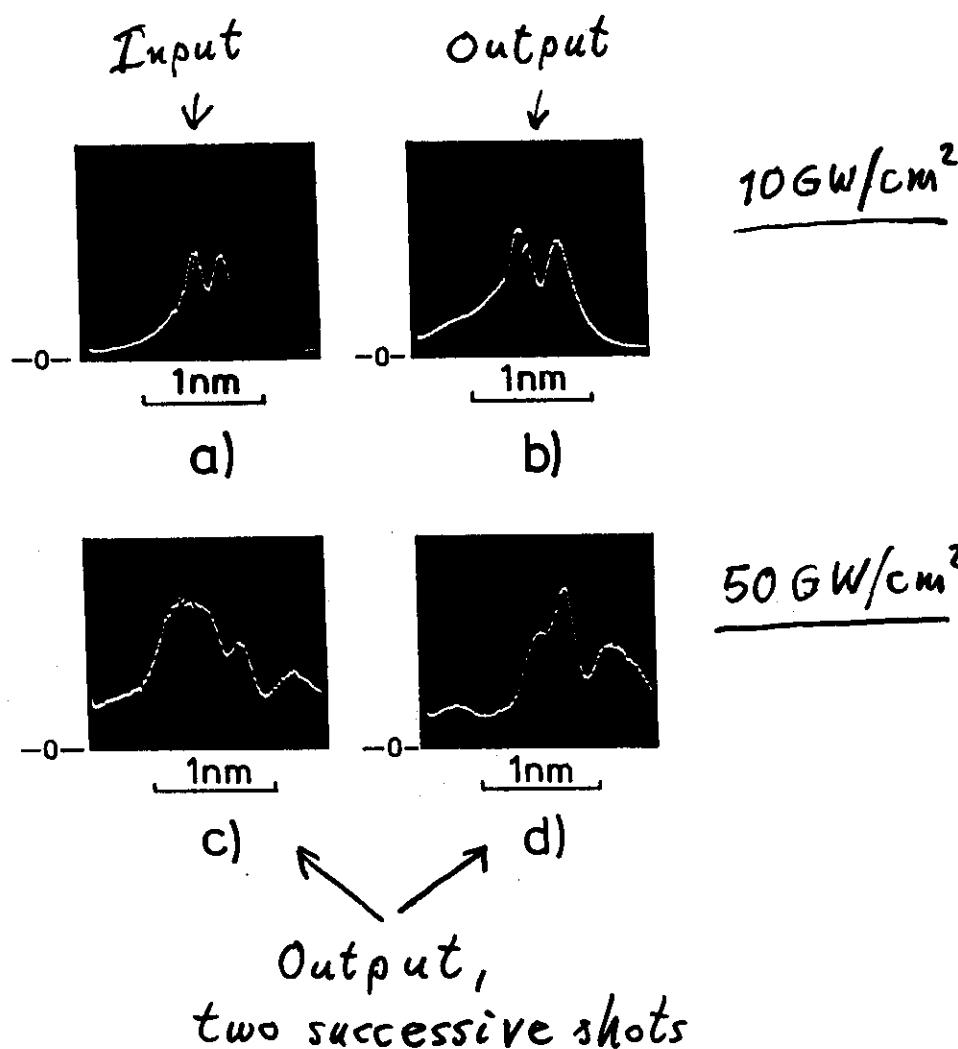
0.11 at $\lambda = 248.5 \text{ nm}$
for ① = LiF ,
② = Quartz

5.12

Attenuation by nonlinear Absorption



UV after 20 μm quartz glass

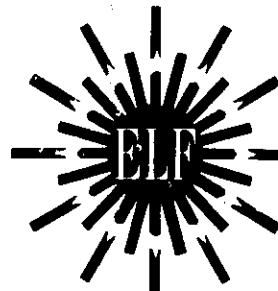


Scientific and Technical Working Group

REPORT



EUROPEAN HIGH PERFORMANCE LASER FACILITY



February 1990

S. Szatmári, G. Kühnle, A. Endoh, F. P. Schäfer, J. Jasny,
Y. W. Lee, J. Jethwa, U. Teubner, G. Kovács

Technical Proposal for the ELF 100 J/100 fs KrF-Laser System SIMBA



EUROPEAN HIGH PERFORMANCE LASER FACILITY

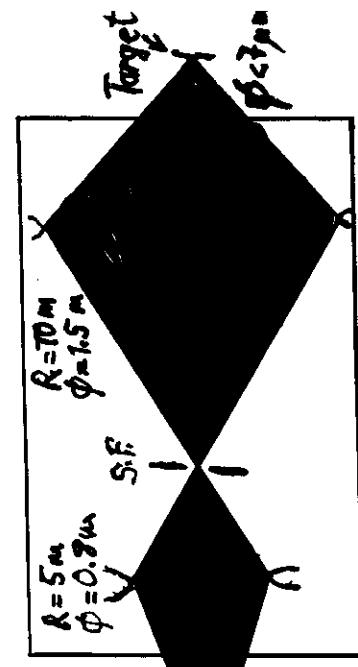


October 1990

European Laser Facility Project

Architecture:

100 J/100 fs KrF Laser



2 e-beam-pumped amplifiers
+ 1:1 imaging with toric mirrors on target

Problems:

1. Windows (CaF_2 , amorphous Teflon)
2. Toric mirrors (Carl Zeiss)
3. Alignment + median. stability
4. Short pulse (10 ns) e-beam T-W.-pumping

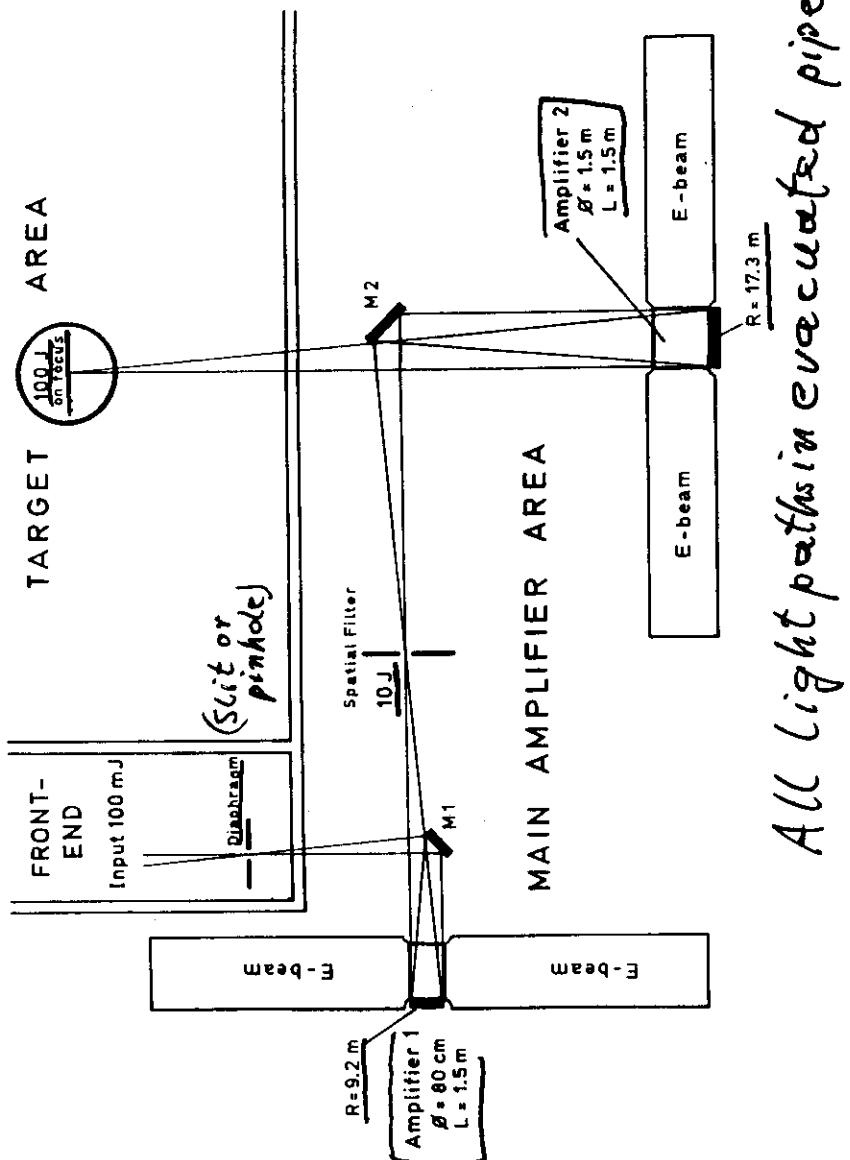


Fig. 21 Geometrical arrangement of the two e-beam-pumped amplifiers.

<3

$$\begin{aligned} R_{1m} &= 9,226 \text{ mm} & R_{1e} &= 9,209 \text{ mm} \\ R_{2m} &= 17,300 \text{ mm} & R_{2e} &= 17,267 \text{ mm} \end{aligned}$$

1:1 Imaging by toric mirrors

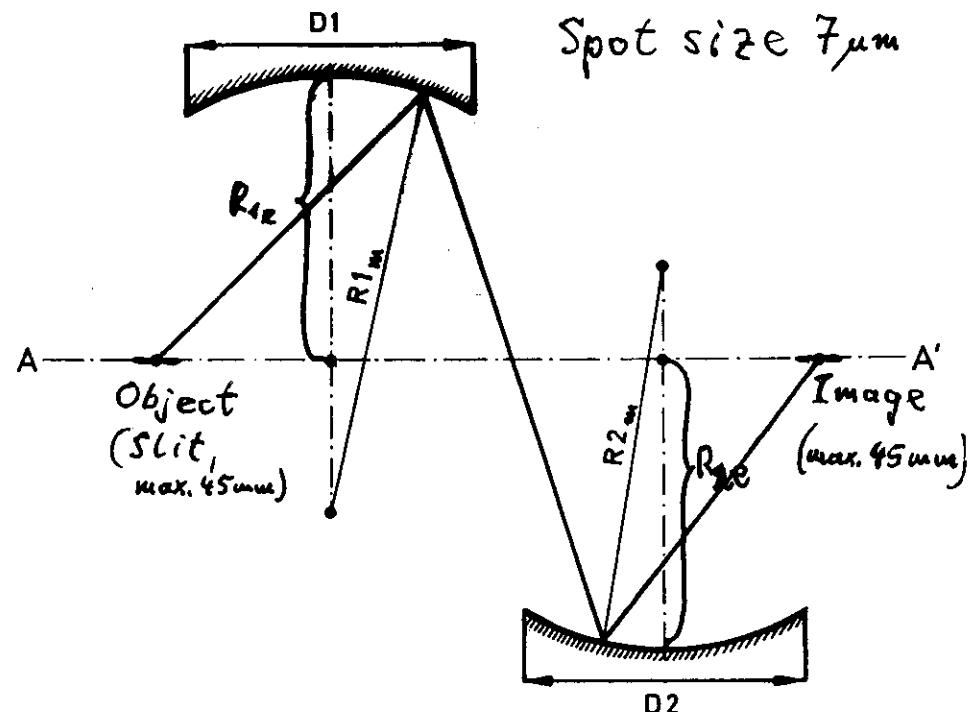


Fig. 20 Two toric mirrors in Z-configuration.

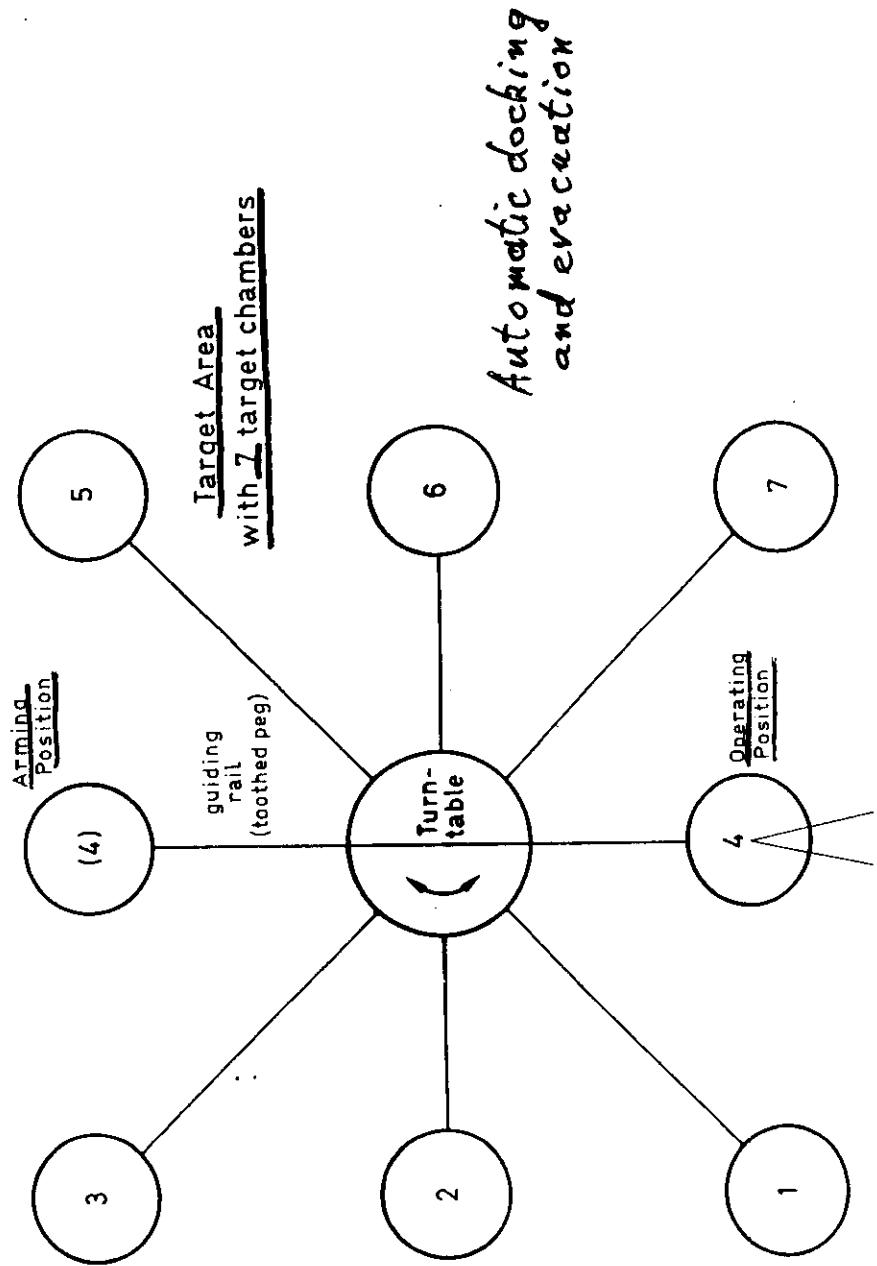


Fig. 23 Lay-out of the target area.

3rd Lecture:

Applications of Ultrashort High Intensity Excimer Laser Pulses

- a) X-Ray Lasers
- b) Chemical Applications

Abstract. One of the most fascinating applications of ultrashort high intensity excimer laser pulses is the realization of a laboratory x-ray laser. After a brief introductory discussion of possible x-ray laser applications the problems connected with the realization of laboratory x-ray lasers are discussed in some detail. It will be shown, why ultrashort pulses in the ultraviolet are most suitable for the most promising x-ray laser schemes and new proposals for their realization are given.

Another broad class of applications of ultrashort excimer laser pulses even at relatively small pulse energies (several mJ) is the study of fast physical and chemical processes in molecules. An example for a pump probe measurement of intramolecular energy transfer in bichromophoric dye molecules is given.

To indicate the extremely wide applicability of ultrashort excimer laser pulses two more examples from different fields will be discussed, namely an application in synthetic organic chemistry and another application in laser mass-spectrometry.

- 3.1 F.P. Schäfer:
On some properties of axicons.
Appl. Phys. B 39, 1-8 (1986)
- 3.2 M. Steyer, F.P. Schäfer, S. Szatmári, G. Kühnle:
Feasibility of a laboratory X-ray laser pumped by ultrashort UV laser pulses.
Appl. Phys. B 50, 265-273 (1990)
- 3.3 G. Kühnle, F.P. Schäfer, S. Szatmári, G.D. Tsakiris:
X-Ray production by irradiation of solid targets with sub-picosecond excimer laser pulses.
Appl. Phys. B 47, 361-366 (1988)
- 3.4 N.P. Ernsting, M. Kaschke, J. Kleinschmidt, K.H. Drexhage, V. Huth:
Sub-picosecond time-resolved intramolecular electronic energy transfer in bichromophoric rhodamine dyes in solution.
Chem. Phys. 122, 431-442 (1988)
- 3.5 D. Plaas, F.P. Schäfer:
Laser photochemistry of aromatic substituted cyclobutanes and cyclobutenes.
Chem. Phys. Lett. 131, 528-533 (1986)

X - Ray Laser Applications

①

X-ray microscopy

(Schmahl, Göttingen)

Advantages: high resolution,
operation in air,
samples up to $1\mu\text{m}$ thick,
living matter unstained.

②

Micro lithography

for semiconductor industry

③

Structure of micro-crystals

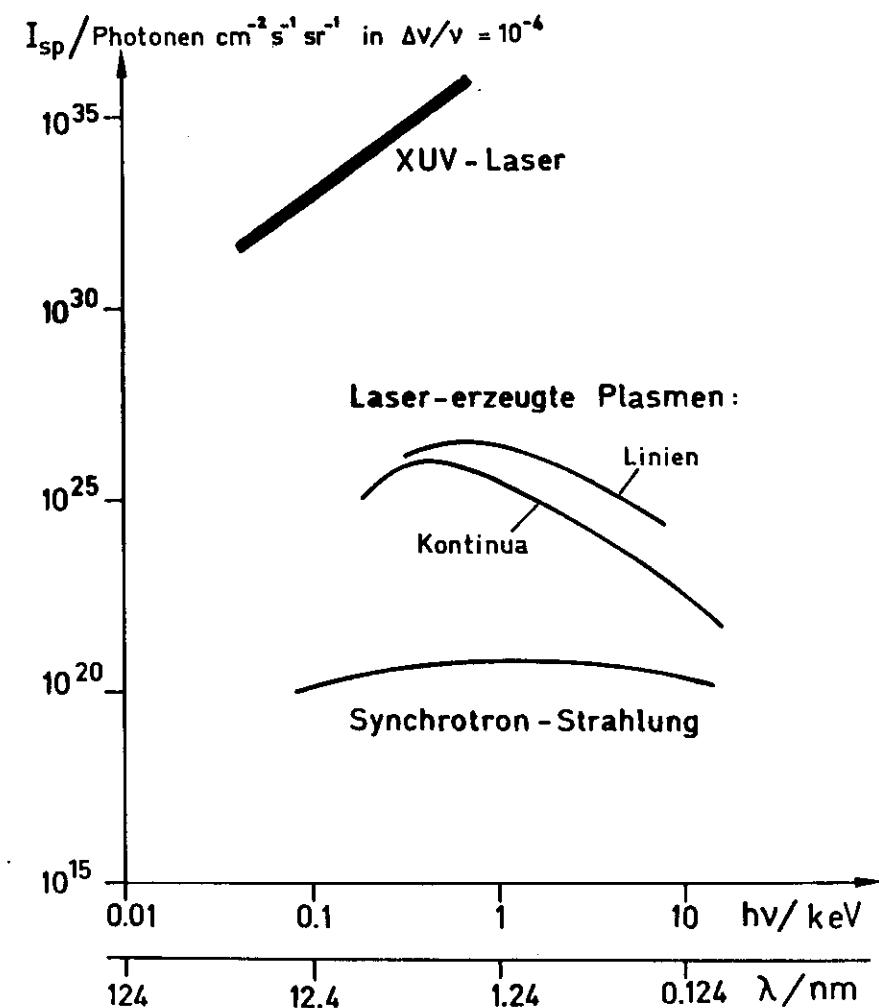
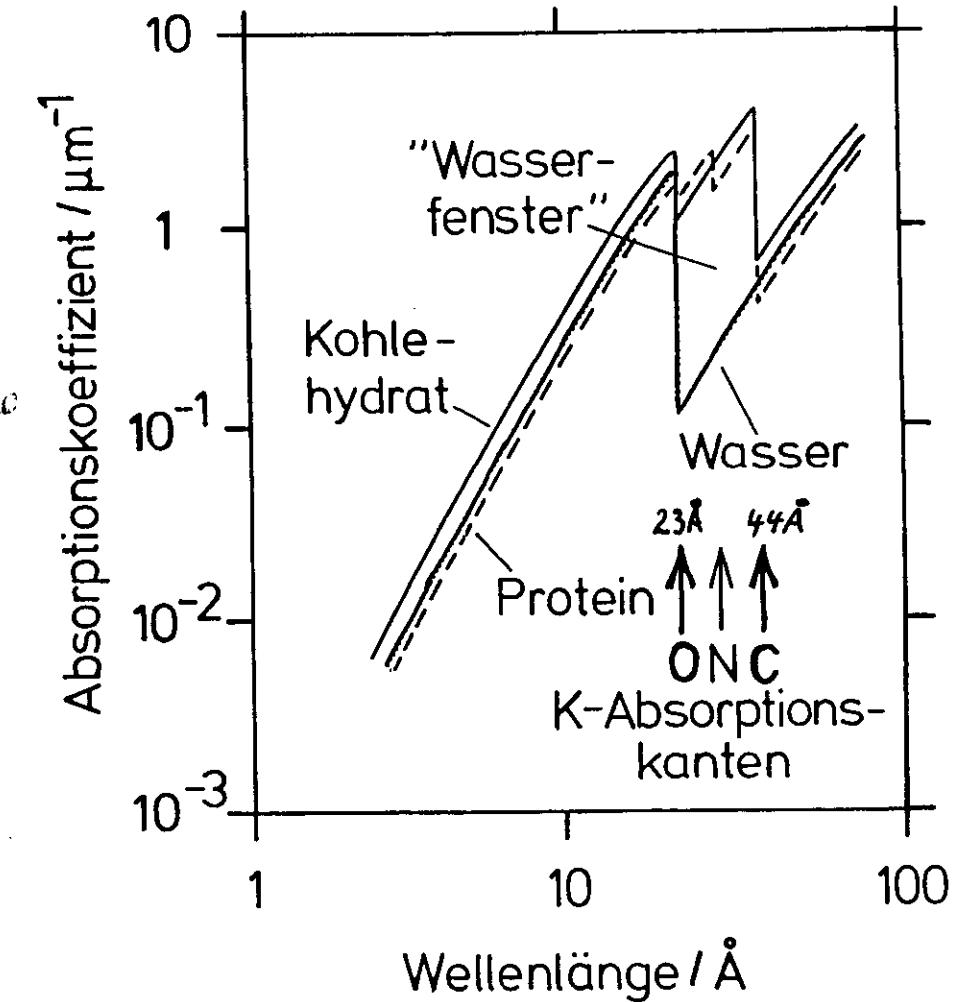
of biological material

④

Shortest laser pulses (< 1 fs)

Comparison of X-ray sources

Vergleich von Röntgenquellen

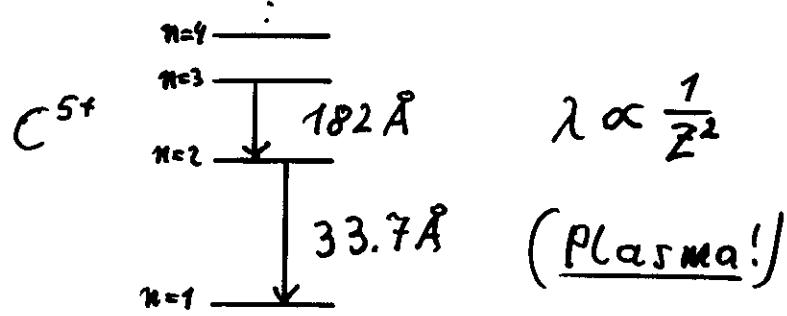


(Quelle: M. H. Key, Nature, 316 (1985) 314)

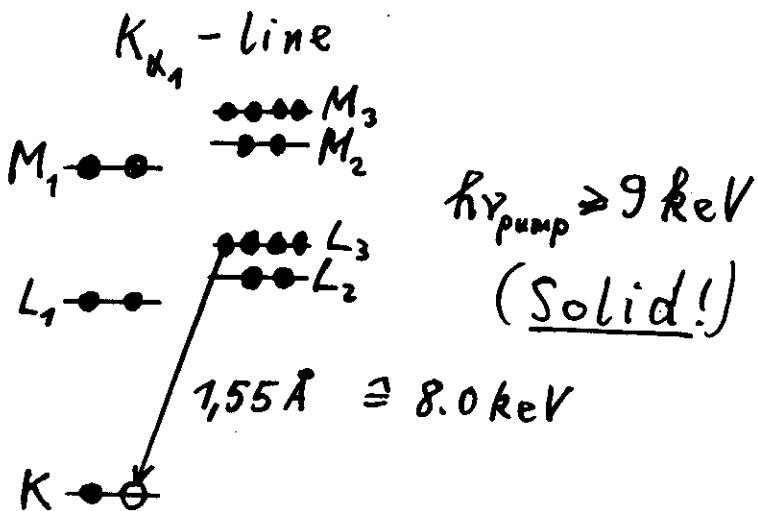
Problem 1

Active medium for lasers in the X-ray region?

⇒ a) "optical" transitions of highly ionized atoms, e.g. H-like ions:



⇒ b) inner-shell transitions after X-ray pumping
e.g. K_{α} -fluorescence of Cu



27

How to build a laser with no absorption-free mirrors available

⇒ Laser without resonator (ASE-Laser)



aspect ratio: d/L } optimum ϕ : $d \approx \sqrt{\lambda \cdot L}$
diffraction limit: λ/d } $\lambda = 10\text{\AA}$, $L = 5\text{mm} \Rightarrow d \approx 3\mu\text{m}$

Laser threshold: one stim. em. per passage
 $\Rightarrow \delta \cdot N^* \cdot L = 1$

"real laser": Gain $G = \exp(\delta \cdot N^* \cdot L) > 1000$

Transit time L/c (16.7 ps for $L = 5\text{mm}$)

Problem 3

How to obtain an inversion?

⇒ Various schemes proposed:

- a) collisional excitation scheme of visible ion lasers shifted to X-ray region in highly charged ions
 - b) recombination of fully stripped atoms to form excited H-like atoms
 - c) recombination in ions with closed shells
 - d) X-ray fluorescence after pumping with ionizing radiation
 - e) X-ray fluorescence after selective excitation of atoms
- ...

Problem 4

How much pump power is needed?

⇒ Example: Cu K_{α1}-line

Cu-wiretarget, $10\mu\text{m} \varnothing$, 5 mm long

Gain: $e^{0.4\pi L} > 1000$

$$\delta_m = \frac{\lambda^2}{4\pi^2 \Delta\nu T_p} \quad \lambda = 1.54\text{\AA} \quad \Delta\nu = 5 \cdot 10^{14}\text{Hz}, T_p = 15\text{fs}$$

$$\Rightarrow \delta_m = 8 \cdot 10^{-18} \text{cm}^2$$

Inversion: $N^* > 1.7 \cdot 10^{18} \text{cm}^{-3}$

total number of excited Cu-atoms:

$$n_{exc} = N^* \cdot \pi \cdot \frac{d^2}{4} \cdot L$$

$$\text{Inversion power } P = n_{exc} \cdot h\nu / \epsilon \Rightarrow \underline{26\text{TW}}$$

Target irradiation intensity:

$$I = P / A \Rightarrow \underline{\underline{1.6 \cdot 10^{16} \text{W/cm}^2}}$$

Problem 5

How much pump **energy** is needed?

⇒ Case a) whole-target irradiation

$$26\text{TW} \cdot 15\text{ps} = 390\text{J}$$

photon-transit time = laser pulse duration

⇒ Case b) travelling-wave excitation

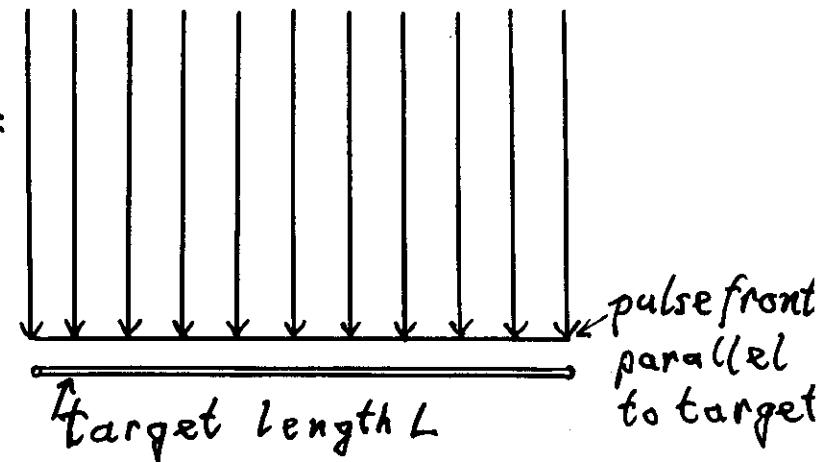
$$26\text{TW} \cdot 100\text{fs} = 2.6\text{J}$$

laser pulse duration at any
irradiated target spot

Pumping of X-ray Lasers

a) conventional Line focus

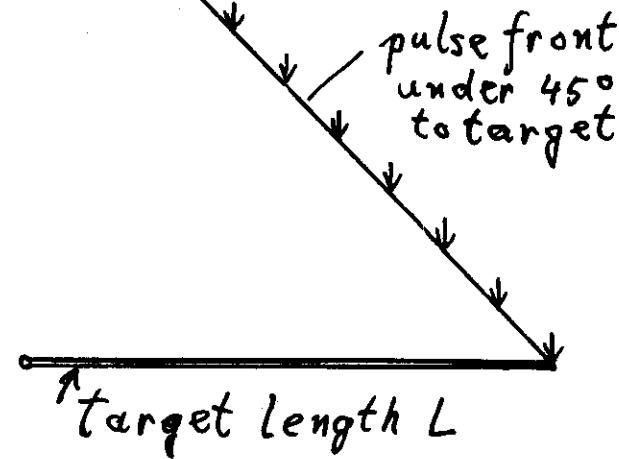
for $L = 10\text{mm}$:
 $T_{p,\min} = 33\text{ps}$



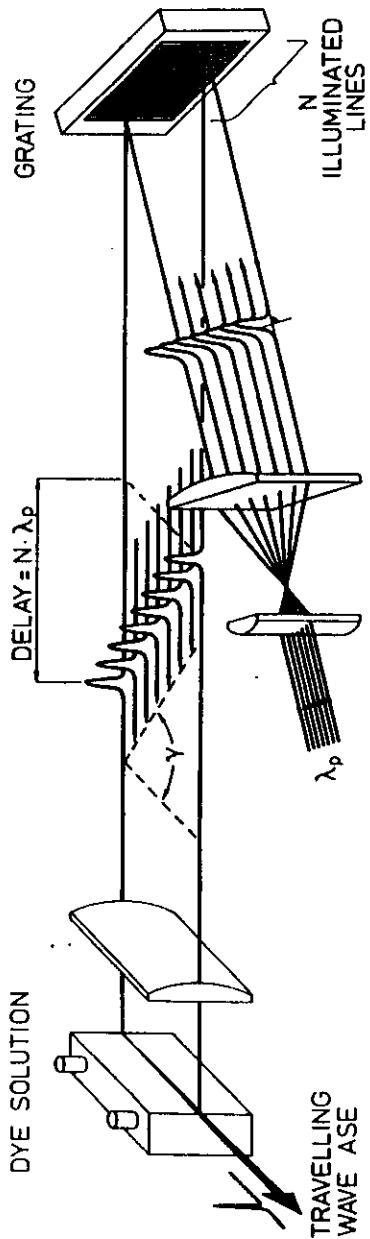
b) travelling-wave pumping

$T_{p,\min} > T_{exc}$
e.g. $T = 100\text{fs}$
 $L = 10\text{mm}$:
330 times
less energy
needed!

23



Principle of travelling-wave pumping



Szatmári/Köhne/Simon: Pulse compression and travelling wave excitation scheme ...

30

Problem 6

Pump Sources for X-Ray Lasers?

1. Nuclear explosions } not for laboratory X-Ray lasers
2. Particle beams
3. High power lasers

System under construction

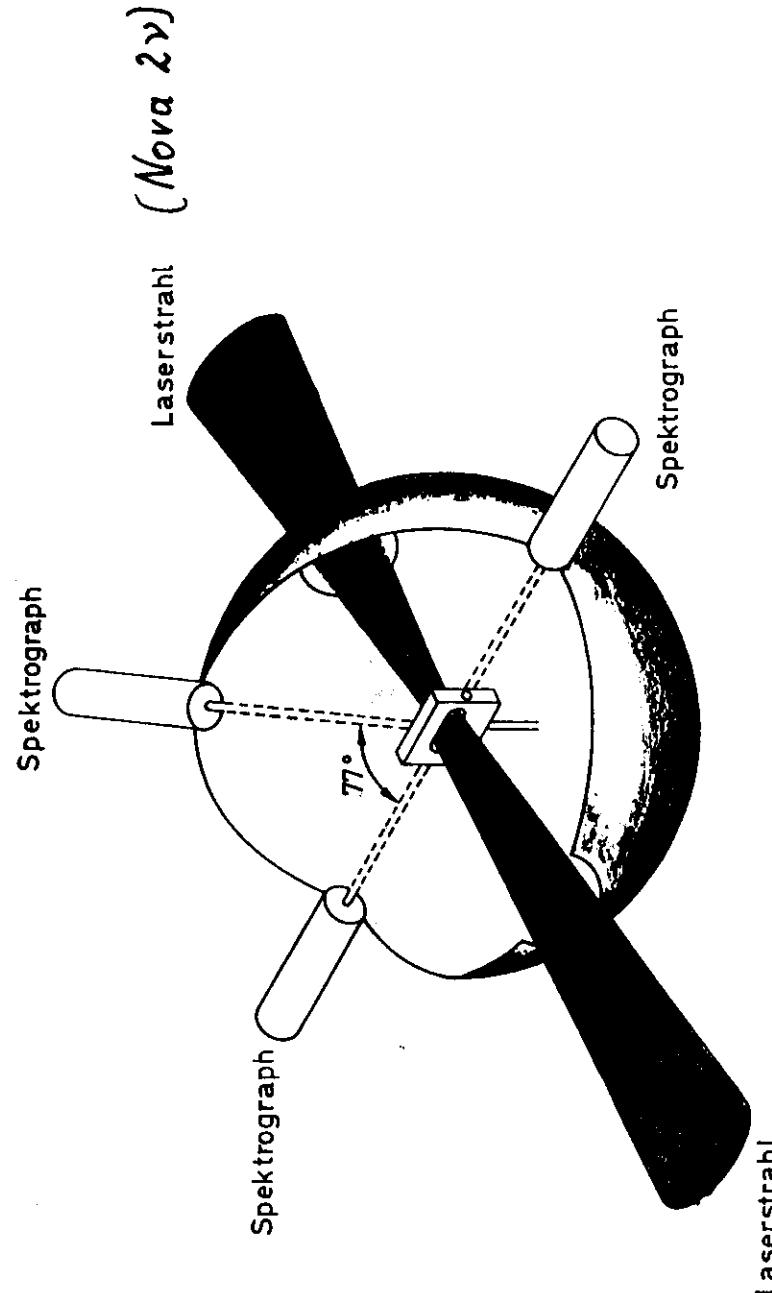
Laser: KrF₂-3-stage amplif., discharge-pumped
100fs, 50mJ \Rightarrow 1J (248nm)

Focussing: Travelling-wave excitation
using special axicon optics

Target: Carbon fiber, $7\mu\text{m}$ ϕ , 5mm length

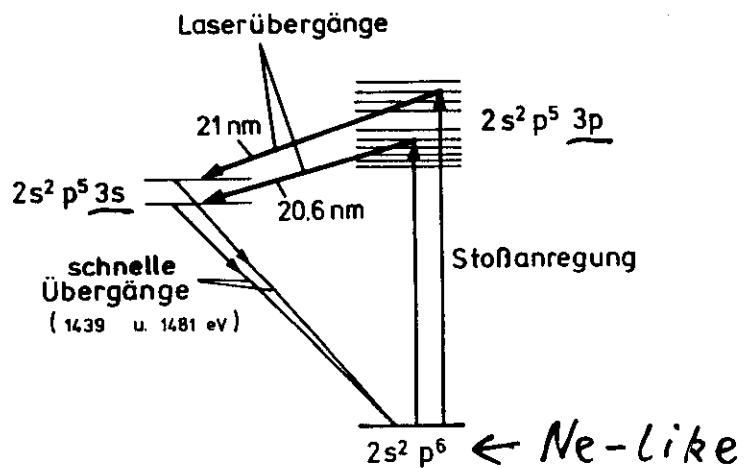
Goal: Recombination ASE laser using
 $2 \rightarrow 1$ transition in C⁵⁺ at 33.7 Å
Efficiency $10^{-9} (\cong 100\mu\text{J}) \cong 1.7 \cdot 10^{12} \frac{\text{photons}}{\text{pulse}} \cong > 100\text{MW}$

w ~



Characteristics of the LLNL - Se⁺²⁴ Laser

Energie-Niveaus Se²⁴⁺



Pump laser: $\lambda = 532 \text{ nm}$
 $E = 2 \text{ kJ}$
 $\tau = 400 \text{ ps}$ } $P = 5 \text{ TW}$

Target: Se evaporated on formvar foil
 $d = 75 \text{ nm}$, $w = 200 \mu\text{m}$, $L \leq 20 \text{ mm}$

Signal: $\lambda = 20.6 \text{ nm}, 21 \text{ nm}$
 $\tau_p = 200 \text{ ps (cw)}$
 $P = 1 \text{ kW (1 MW)}$ } $\eta = 10^{-10} (10^{-7})$
Gain: $700 \text{ (} g_o \approx 5.5 \text{ cm}^{-1} \text{)}$

Plasma: $n_e \approx 10^{21} \text{ cm}^{-3}$
 $T_e \approx 5 \cdot 10^6 \text{ K}$

Other Realized "X-Ray Lasers"

λ [nm]	Laser-Ion	Labor/Laser
28.6		
24.7		
23.6	Ge ²²⁺	<u>NRL/Pharos III</u>
23.2	Ne-like	
19.6		
22.1	Cu ¹⁹⁺	<u>NRL/Pharos III</u>
26.29	Ne-like	
22.03		
20.96	Se ²⁴⁺	<u>LLNL/Nova</u>
20.63	Ne-like	
18.2	C ⁵⁺ H-like	<u>RAL/Vulcan, PPL</u>
15.5	Y ²⁹⁺	<u>LLNL/Nova</u>
13.94	Ne-like	
13.27		
13.1	Mo ³²⁺	<u>LLNL/Nova</u>
10.64	Ne-like	
10.57	Al ¹⁰⁺ Li-like	UPS
8.1	F ⁸⁺ H-like	<u>RAL/Vulcan</u>
7.1		
6.85	Eu ³⁵⁺	<u>LLNL/Nova</u>
	Ni-like	

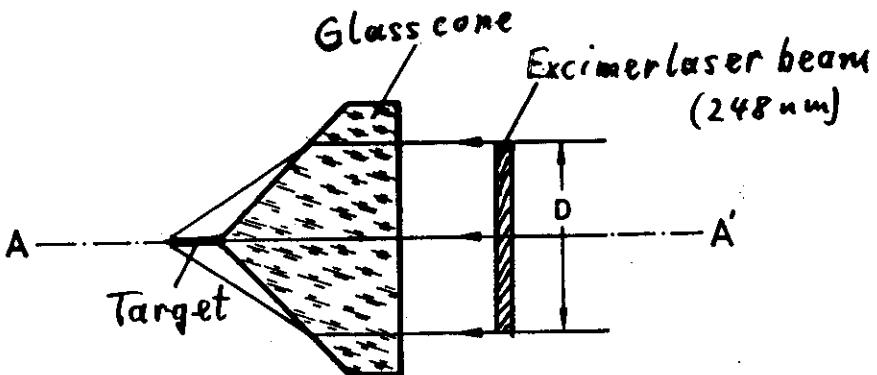


Fig. 1

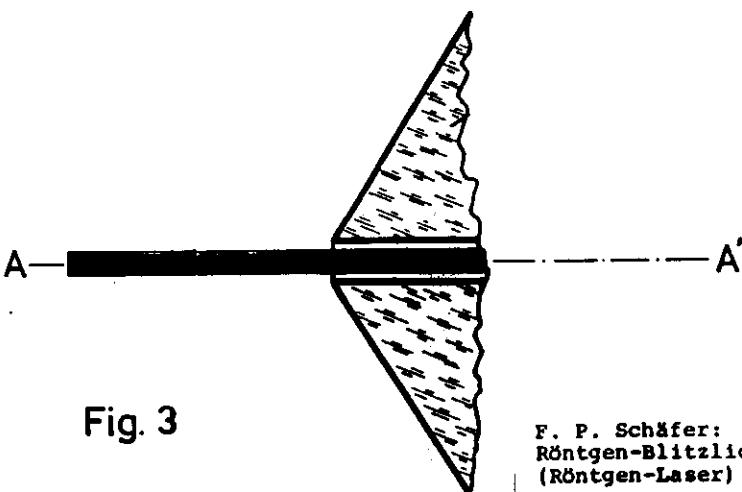
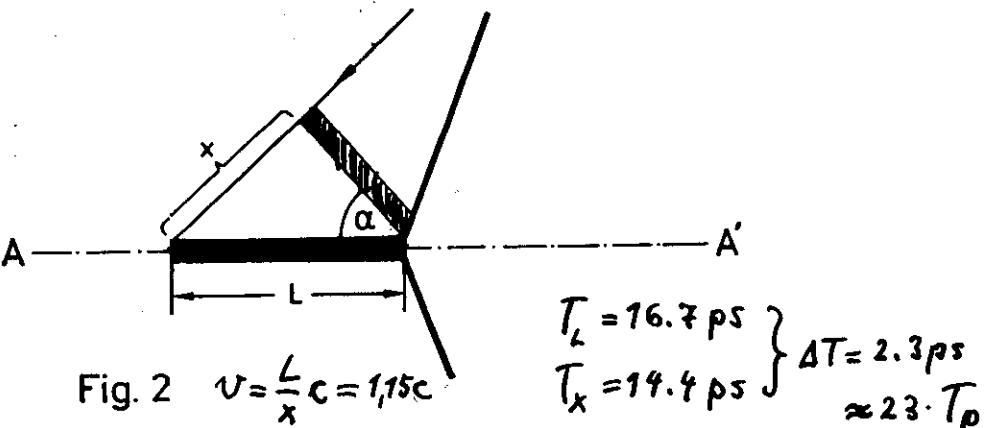
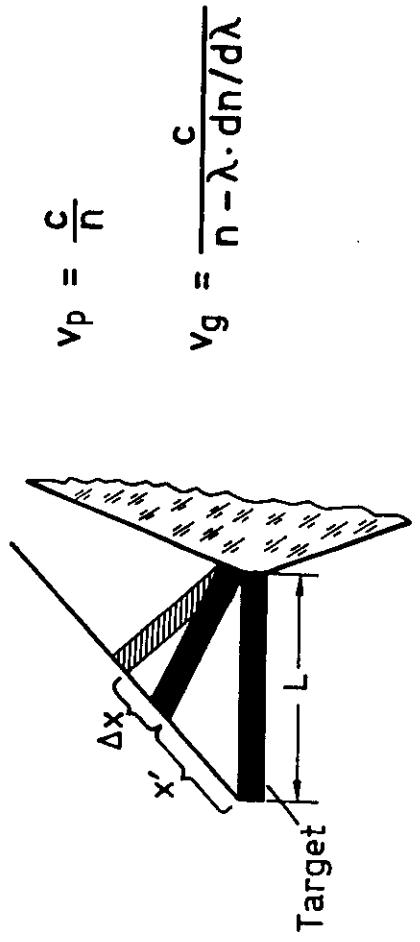


Fig. 3

F. P. Schäfer:
Röntgen-Blitzlichtquelle
(Röntgen-Laser)

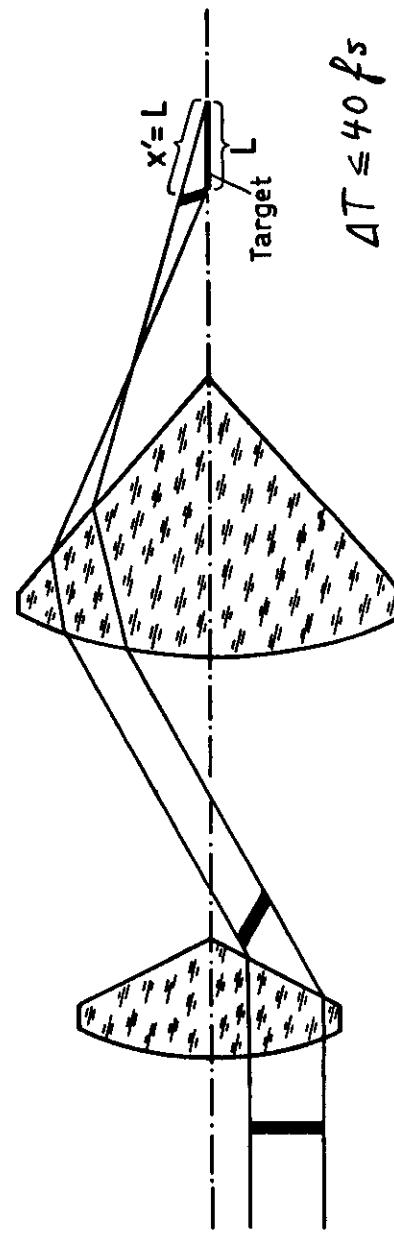
Schiefe Impulsfront (Gruppengeschwindigkeit)

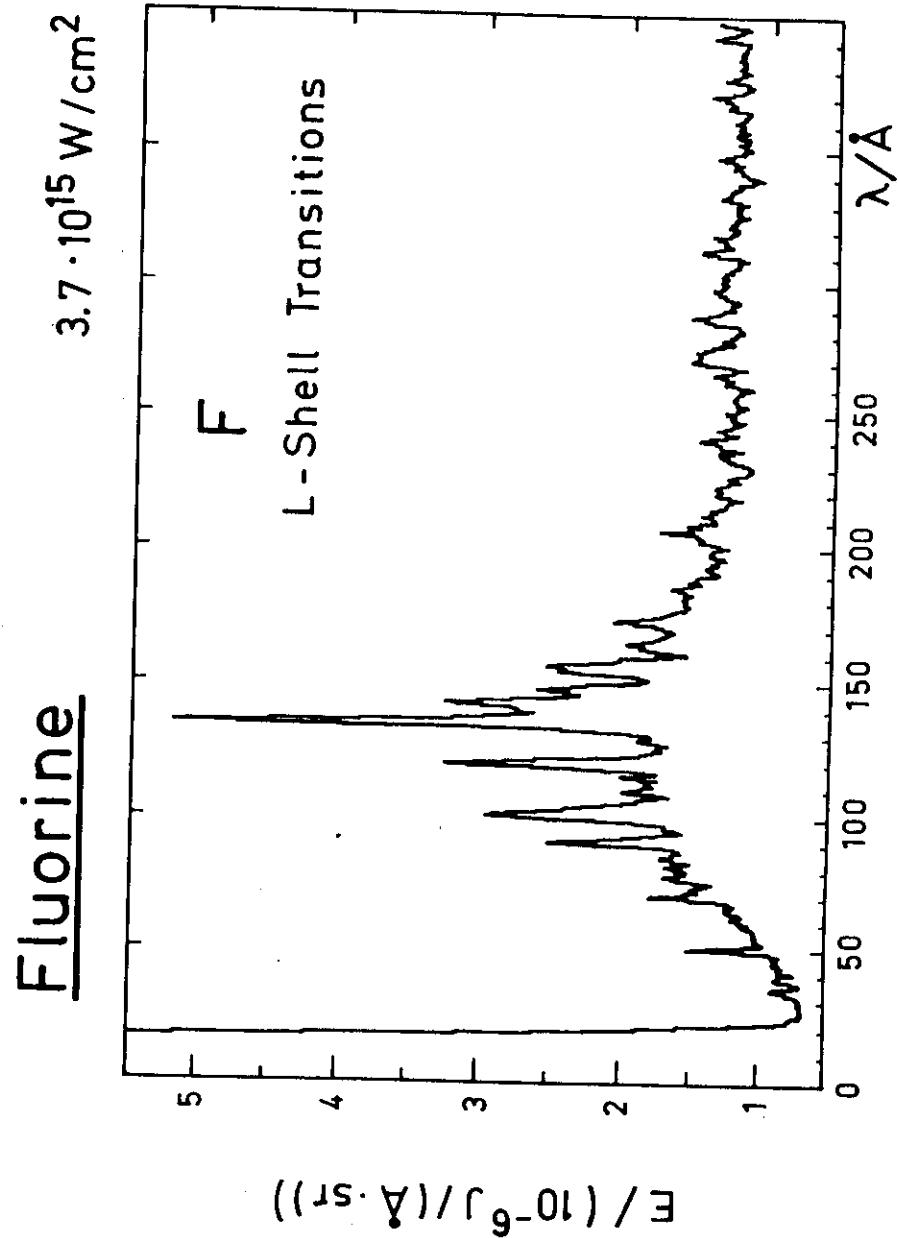
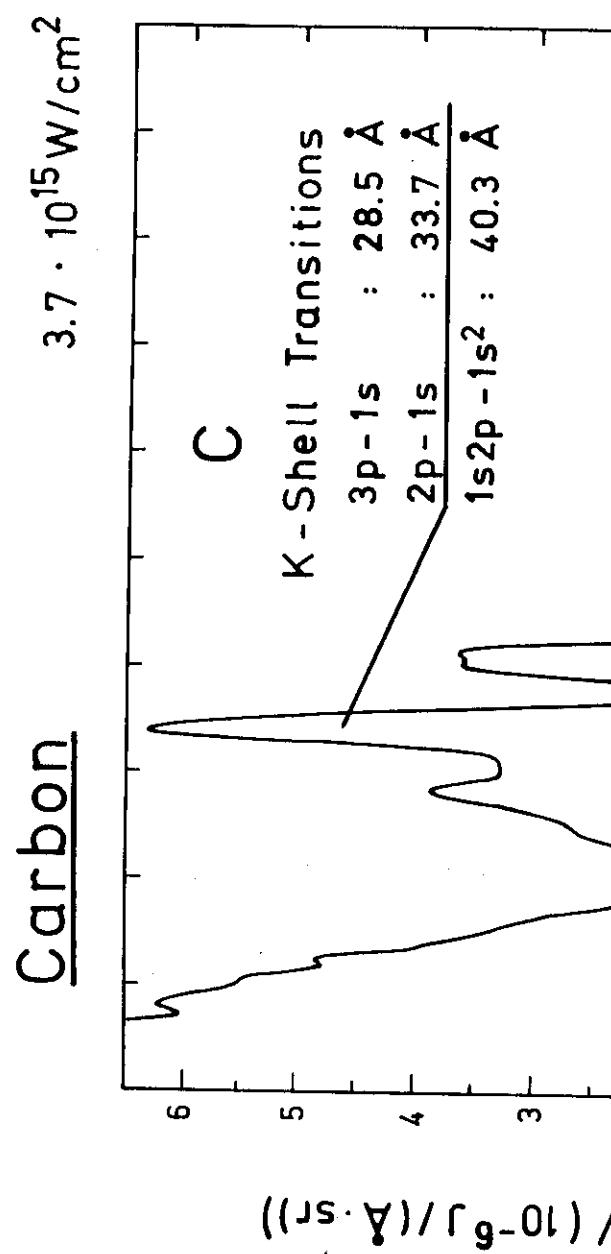


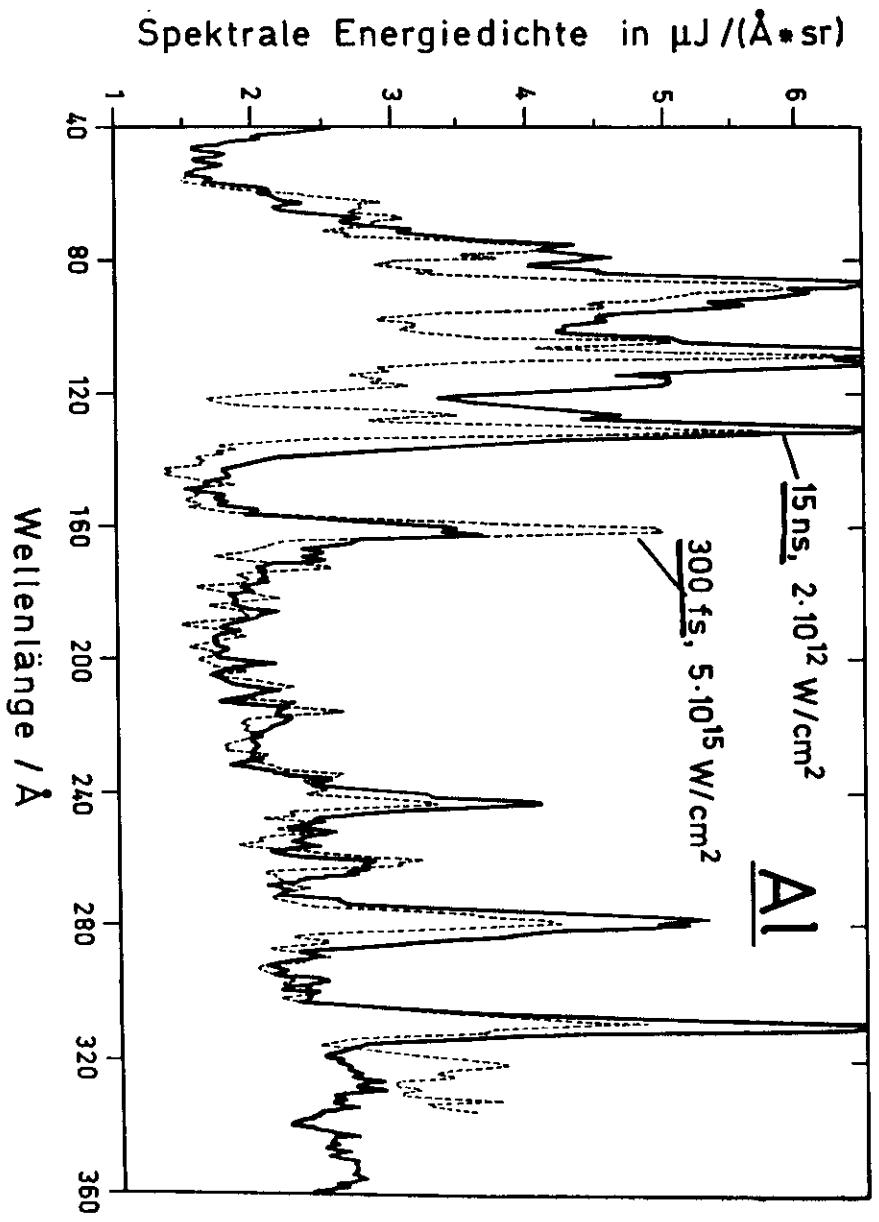
$$\text{Wanderwellengeschwindigkeit } v = \frac{L}{x'} c$$

35

Zeit-korrigierte Pump-Optik

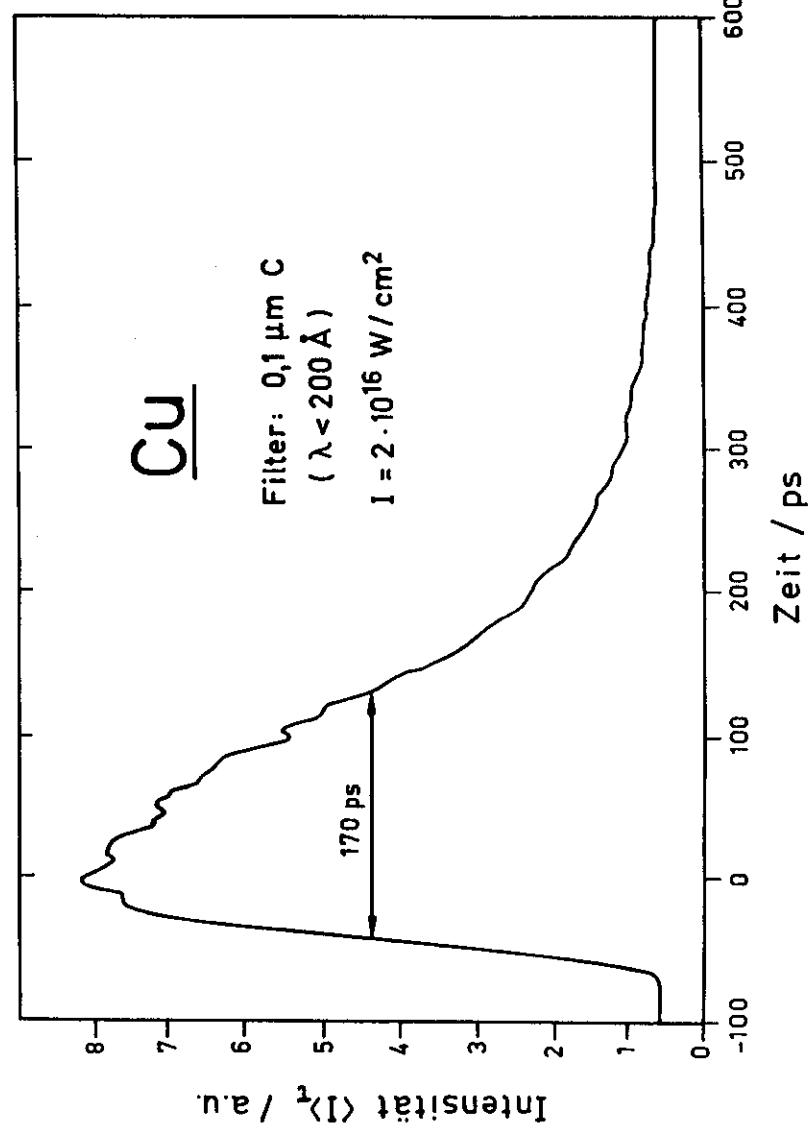




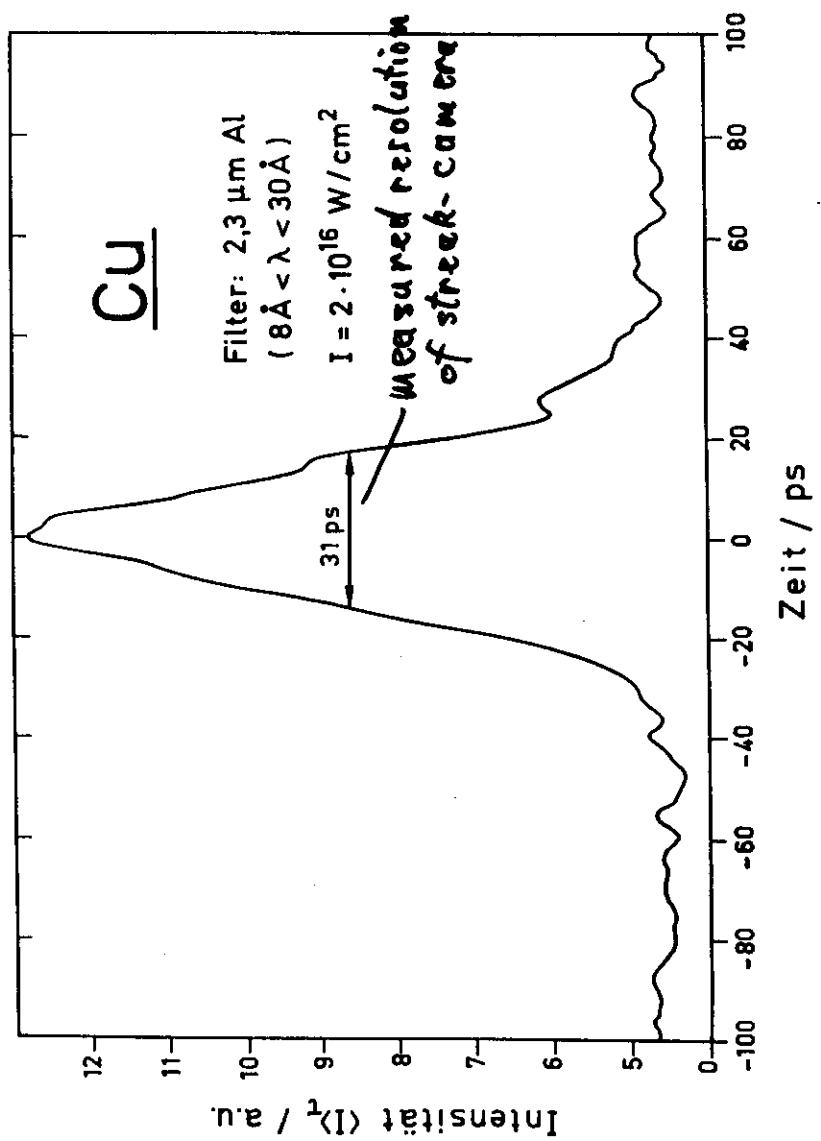


Comparison of X-ray spectra excited by long or short pulses

ns

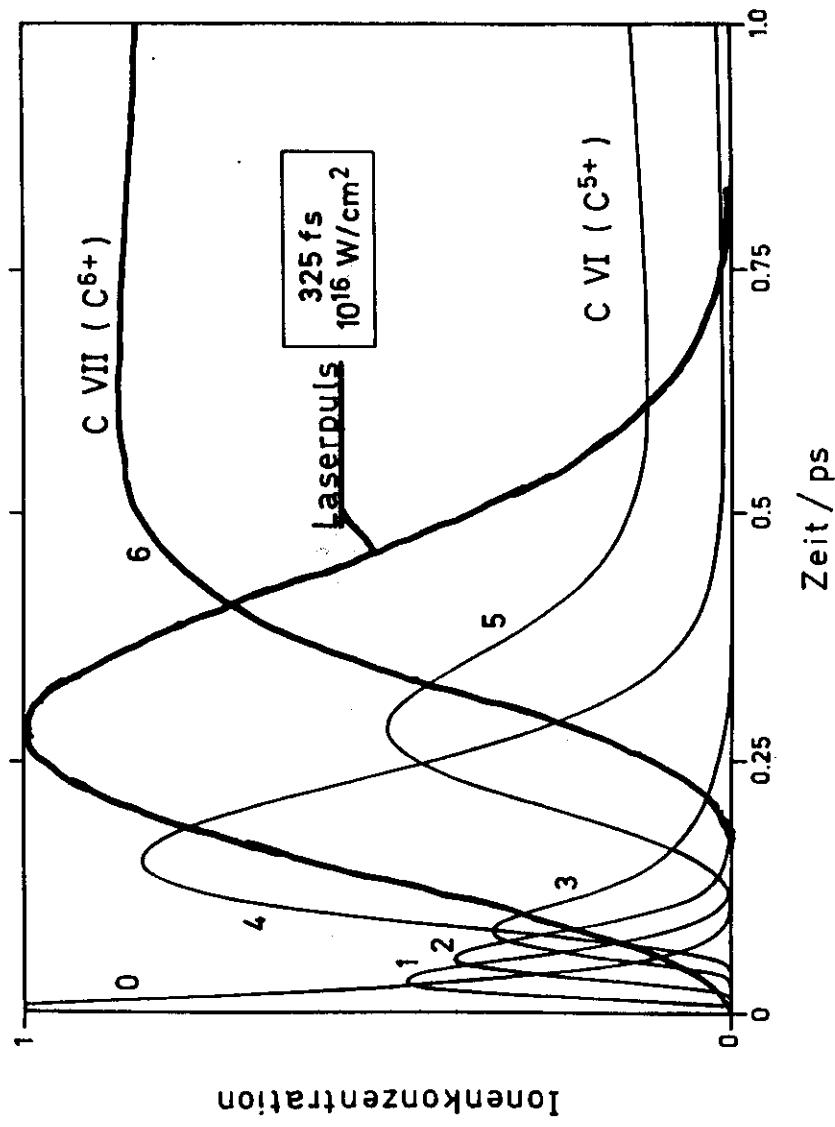


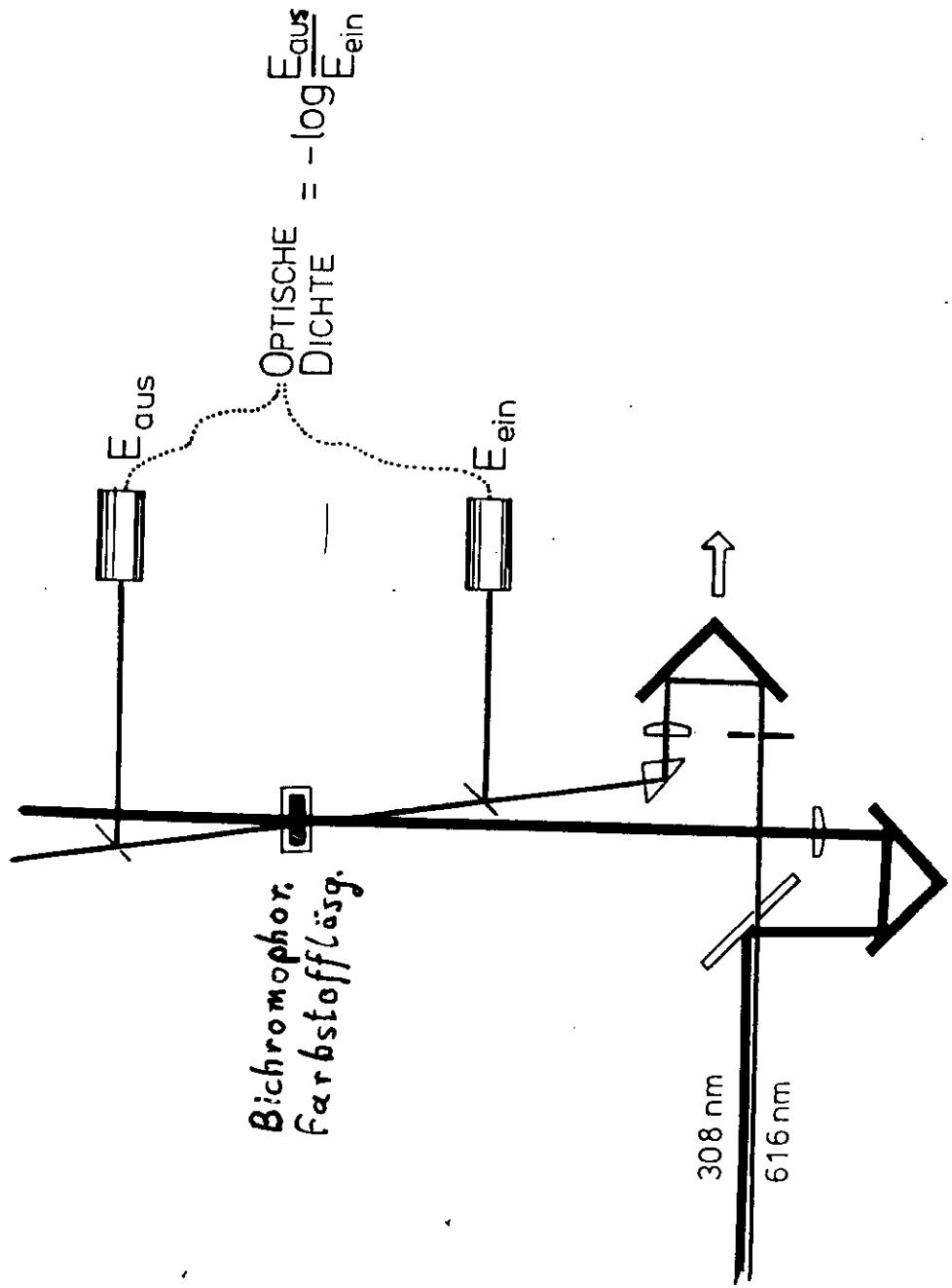
X-ray streak-camera record



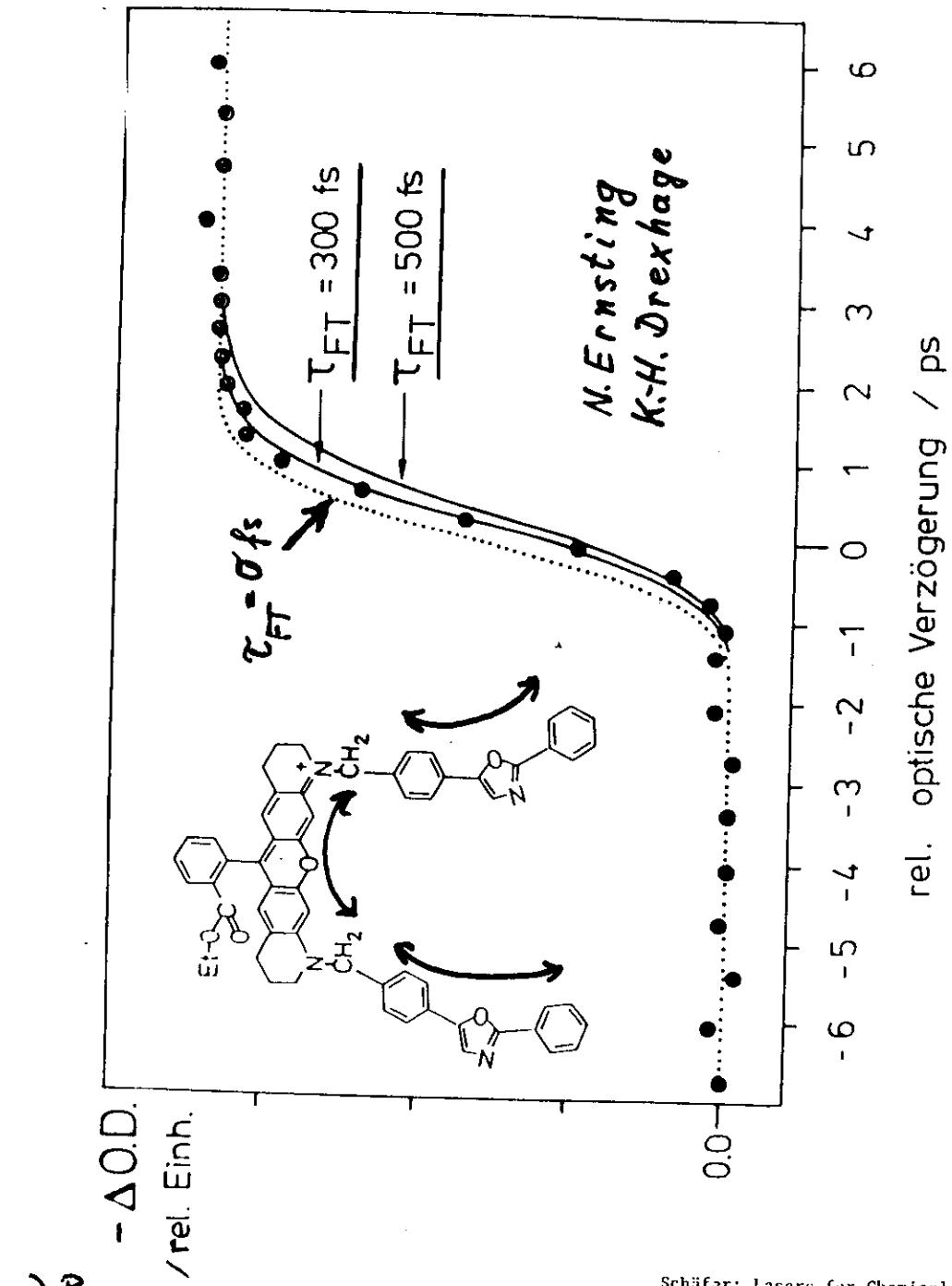
37

Temporal development of ionization (Keldysh model)





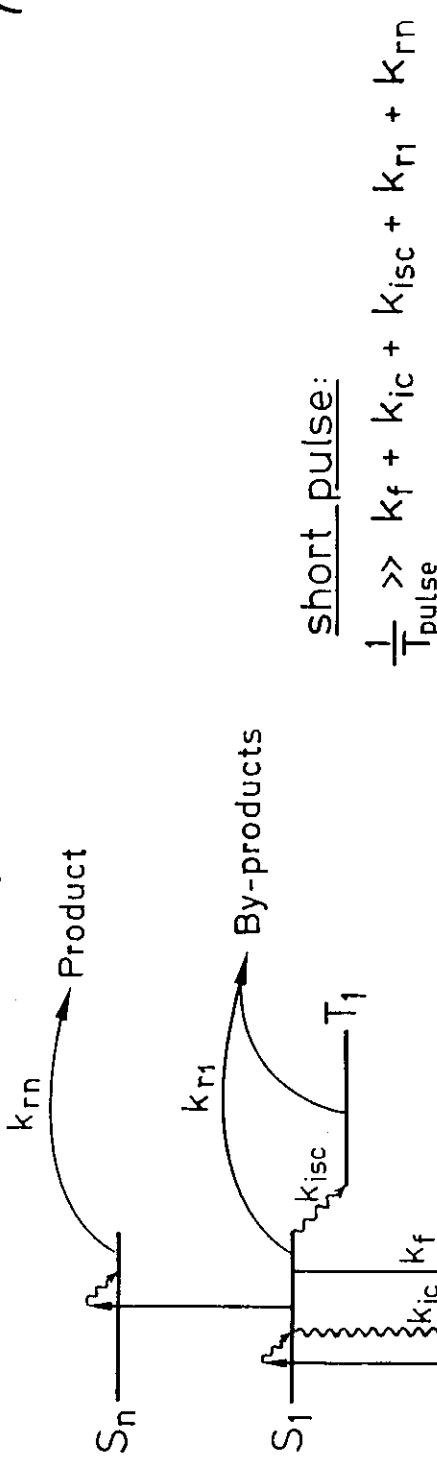
Schäfer: Lasers for Chemical Applications



Schäfer: Lasers for Chemical Applications

Fig. 21

Advantages of pulsed irradiation in photochemistry



Advantages of short pulses:

1. Higher ratio of products over by-products
2. Higher quantum yield
3. Higher chemical yield
4. Higher intensities ($\text{intensity}_{\text{product}} \sim I^2$)

9

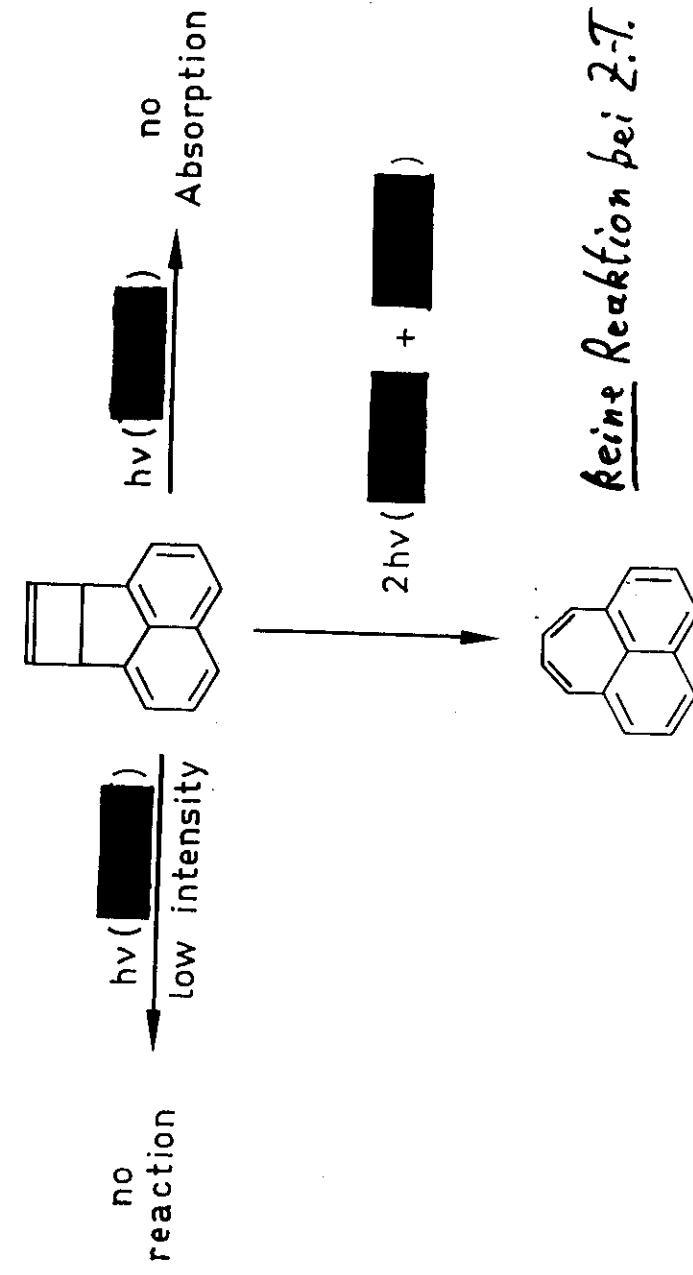
chäfer: Lasers for Chemical Applications.

Fig. 13

12.16

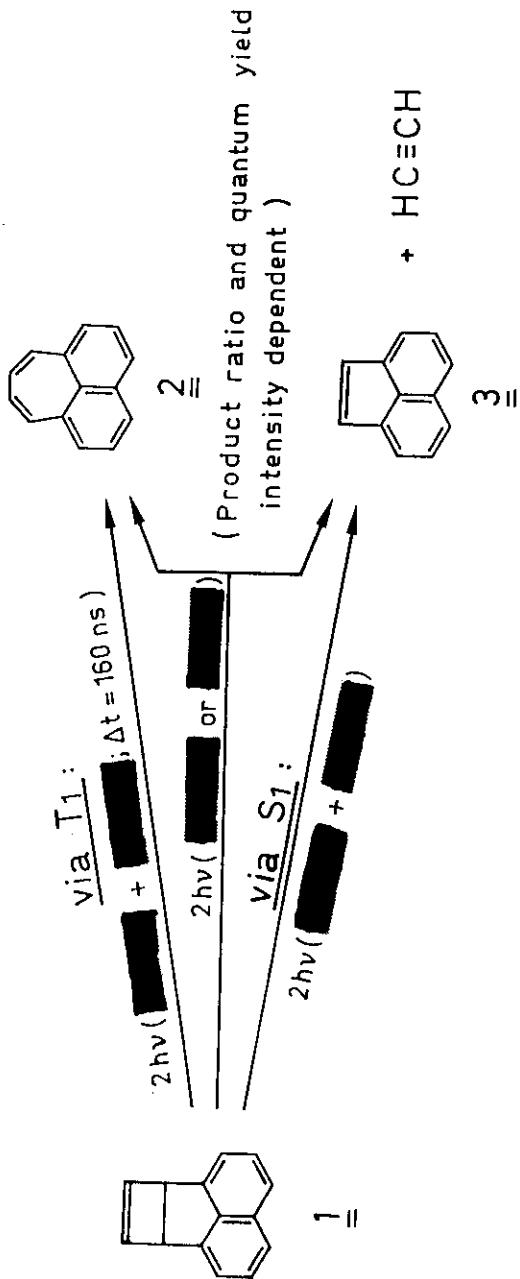
2-Photon - (2-Step-) Reaction

organic glass / 77K



(12.20)

Liquid Solution at Room Temperature



Schäfer: Lasers for Chemical Applications.

90

