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TRAINING COLLEGE ON PHYSICS AND CHARACTERIZATION OF LASERS AND OPTICAL FIBRES

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DYE LASERS

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Dye Lasers

I. General Characteristics

Active medium: liquid solution of a dye
  Solvent: ethanol, ethylene glycol, water, etc.
  Dye: Polyphenyl 1, Rhodamine 6G, HITC, etc.
Working range: 300 - 1800 nm; best efficiency 400 - 900 nm
Tunability of a single dye: 50 nm
Pulse duration: 10 fs - continuous
Pump source: flash lamp, excimer laser, Ar-ion laser, etc.
Power, pulsed: typically MW
Power, continuous: several W

II. Organic Dyes

Dye = organic compound with strong absorption for visible radiation (or near UV or near IR)

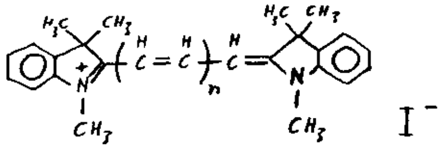
"strong" means: molar decadic extinction coefficient

$$\epsilon > 10^4 \text{ l/mole cm}$$

definition of ϵ: $I = I_0 \cdot 10^{-\epsilon c l}$

with I_0 light intensity before sample
I light intensity behind sample
c concentration of dye
l length of sample (dye cell)

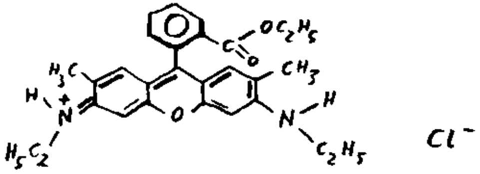
Strong absorption in organic compounds is due to extended π-electron systems, symbolized by alternating single and double bonds:



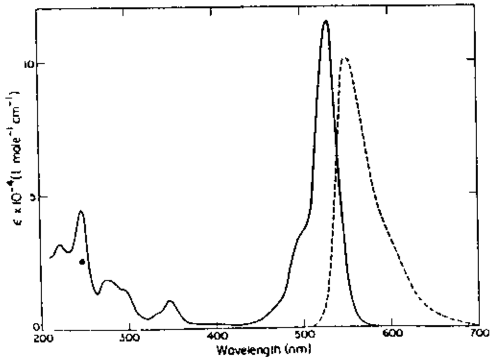
n	name	λ_{abs}
1	HIC	550 nm
2	HIDC	650 nm
3	HITC	750 nm

In cyanine dyes the absorption shifts linearly to longer wavelengths with increasing chain length; about 100 nm per each additional double bond (electron gas model of organic dyes).

A dye related to simple cyanines is rhodamine 6G:



Absorption and fluorescence of rhodamine 6G in ethanol:



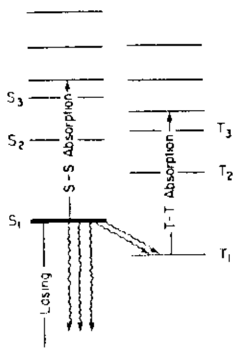
Many dyes are capable of emitting light (fluorescence). This property is essential for their use as laser dye. Nonradiative transitions compete with the emission of a photon. Therefore the quantum yield of fluorescence η_{fl} varies many orders of magnitude among dyes.

Definition: $\eta_{fl} = \frac{\text{photons emitted}}{\text{photons absorbed}}$

In efficient laser dyes: $\eta_{fl} > 0.1$.

Fluorescence involves the same electronic levels as the longest-wavelength absorption band: singulett levels S_0 and S_1. Yet it is red-shifted (Stokes-shift) due to solvent reorientation after excitation. The width of absorption and fluorescence bands is caused by vibronic sublevels and solvent effects. The width of the fluorescence band is responsible for the wide tuning range of dye lasers and makes possible the generation of ultrashort pulses by mode locking.

Energy level diagram of a dye:



Excited dye molecules can cross over from the S_1-level to a triplet level. Because triplet molecules are relatively long-lived (10^{-6} s in solution) and can absorb laser radiation, intersystem crossing is detrimental to laser action. Only dyes with low triplet yield are efficient laser dyes.

In the absence of stimulated emission (laser action) the radiative (natural) lifetime τ_n of the dye in state S_1 is related to the oscillator strength f of the $S_0 \rightarrow S_1$ absorption band by

$$\frac{1}{\tau_n} = \frac{8\pi^2 e_0^2}{m_0 c_0} n^2 \tilde{\nu}^2 f \; .$$

e_0	charge of electron
m_0	mass of electron
c_0	velocity of light
n	refractive index of solution
$\tilde{\nu}$	wave number of center of absorption band

Since the f-value is near unity in most dyes, the radiative lifetime τ_n amounts typically to a few nanoseconds. The real decay time τ of state S_1 is modified by the fluorescence quantum yield:

$$\tau = \eta_{fl} \cdot \tau_n$$

In a dye laser it is further reduced by stimulated emission.

III. Pumping Considerations

4-Level laser: population inversion relatively easy to achieve

Problems: spontaneous decay time of upper laser level very short (only a few ns);
population of triplet level detrimental

In order to avoid triplet problem, rise time of pumping radiation has to be much faster than triplet decay (on the order of µs). Therefore pumping by ns-pulses of excimer laser or Q-switched ruby laser is very efficient. Fast rising flashlamps work too.

Continuous (cw) lasers: Dye solution flows fast through pumping region, so that triplet molecules are removed out of lasing volume as fast as they are formed (no build-up).

Dye laser in its simplest form: Cuvette of length L filled with dye solution of concentrations (molecules/cm³) n_0 (ground state) and n_1 (excited state); parallel end windows carrying a mirror each of reflectivity R; no triplet molecules. Laser starts oscillating at wave number $\tilde{\nu}$, if overall gain

$$\exp[-\sigma_a(\tilde{\nu})n_o L]\cdot R\cdot \exp[+\sigma_e(\tilde{\nu})n_1 L] \geq 1 \quad .$$

$\sigma_a(\tilde{\nu})$ cross section for absorption

$\sigma_e(\tilde{\nu})$ cross section for stimulated emission

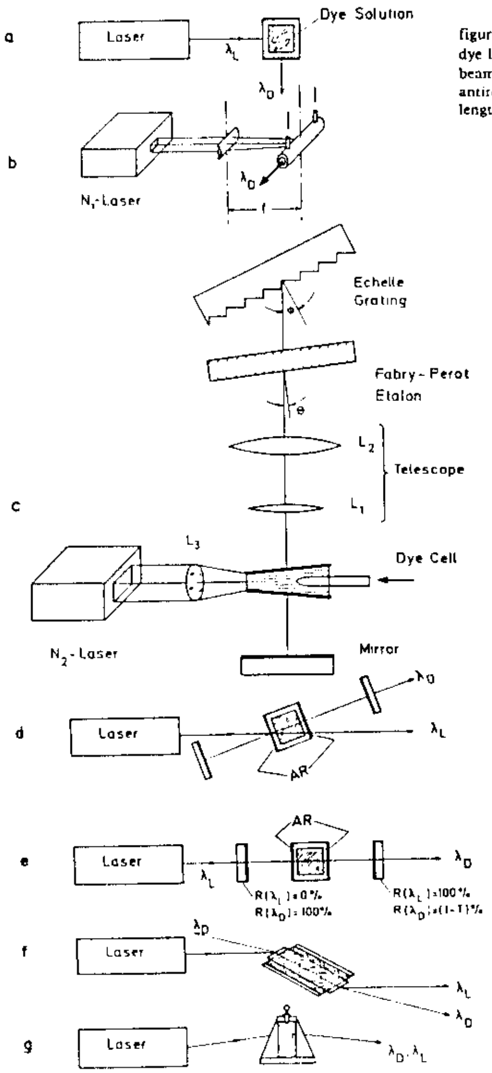
$$\sigma_a = 0.385 \times 10^{-20}\,\epsilon \quad (\sigma_a \text{ in cm}^2,\ \epsilon \text{ in l/mole cm})$$

$$\sigma_e(\tilde{\nu}) = \frac{1}{8\pi c_0 \tilde{\nu}^2}\,\frac{1}{\tau_n}\,F(\tilde{\nu})$$

$F(\tilde{\nu})$ quantum spectrum of fluorescence, normalized according to

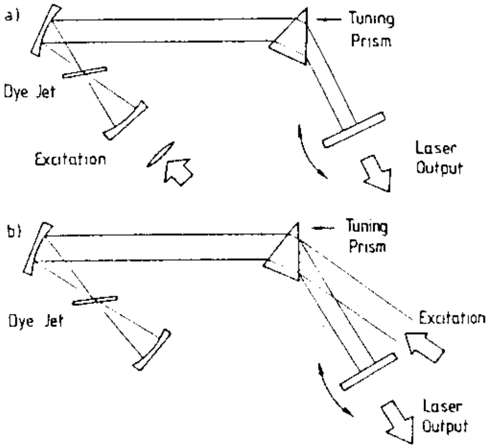
$$\int_{\substack{\text{fluorescence}\\ \text{band}}} F(\tilde{\nu})\,d\tilde{\nu} = 1$$

A large number of pumping arrangements and resonator configurations are in use. For pulsed lasers:

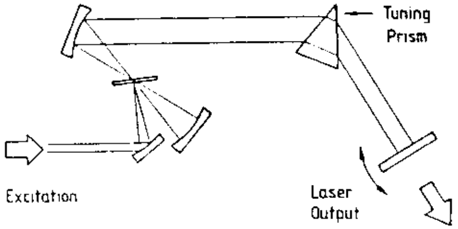


Resonator configurations for laser-pumped dye lasers. (λ_L pumping-laser beam, λ_D dye-laser beam, AR antireflection coating, f focal length of cylindrical lens

In continuous lasers the dye solution is not confined to a cell. It rather moves in form of a jet through the active region. This prevents triplet build-up as well as problems due to thermal inhomogeneities.



Tunable three-element dye laser resonator using reflecting optics. (a) Collinear excitation through end mirror. (b) Collinear excitation through tuning prism



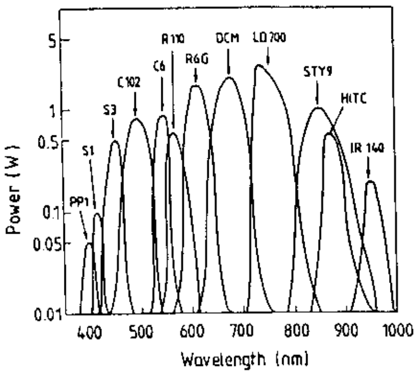
Noncollinear excitation of tunable three-element dye laser resonator

For continuous operation only dyes with minimal internal losses are useful:

Dye	Short form	Solvent	Excitation	References
Polyphenyl 1	PP1	EG	Argon all lines UV	Huffer et al. (1980)
Stilbene 1	S1	EG + ME (10 : 1)	Argon all lines UV	Huffer et al. (1979)
Stilbene 3	S3	EG + ME (10 : 1)	Argon all lines UV	Kuhl et al. (1978)
Coumarin 102	C102	EG + BZ (10 : 1)	Krypton all lines violet	Yarborough (1974)
Coumarin 6	C6	GL + BZ (1 : 1)	Argon 488 nm	Yarborough (1974)
Rhodamine 110	R110	EG	Argon all lines	Yarborough (1974)
Rhodamine 6G	R6G	EG	Argon all lines	Yarborough (1974)
DCM		EG + BZ (3 : 2)	Argon all lines	Marason (1981)
LD700		EG	Krypton all lines red	Johnston et al. (1982)
Styryl 9	STY9	EG + PGC (17 : 3)	Argon all lines	Hoffnagle et al. (1982)
HITC		EG + DMSO (1 : 1)	Krypton all lines red	Romanek et al. (1977)
IR140		EG + DMSO (1 : 1)	Krypton all lines infrared	Leduc and Weisbuch (1978)

Abbreviations: EG: ethylene glycol; ME: methyl alcohol; BZ: benzyl alcohol; GL: glycerol; PGC: propylene glycol carbonate; DMSO: dimethyl sulfoxide

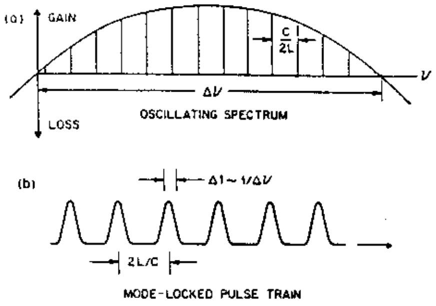
With these dyes the wavelength range 380 nm - 1000 nm is covered. Tuning curves:



Pump power: 3 W in the UV, 4 W in the visible, 2 W in the infrared. Energy conversion efficiency: up to 40 %.

IV. Ultrashort Pulses

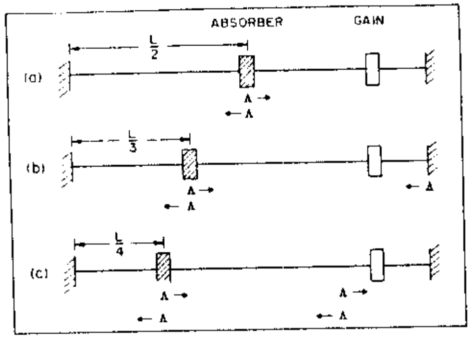
The output of a laser consists generally of a set of resonator modes whose frequencies are spaced by an amount c/2L (c velocity of light, L cavity length). In general the mode phase is randomly fluctuating. By fixing the phase relationship between the modes a periodic train of pulses results.



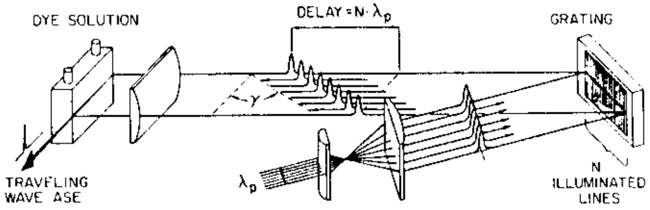
(a) The spectral content of a laser operating in the fundamental transverse mode but in several longitudinal modes. (b) Temporal output of a laser when mode-locked

In case of a single pulse in the optical cavity, the period of the pulse train is T = 2L/c. The width of the pulses is inversely proportional to the width of the oscillating spectrum. Because this extends with dyes over nearly 100 nm, the optical pulses can be as short as 10 fs.

The phase locking of the modes ("mode locking") can be brought about by a saturable absorber - again a dye. This passive mode locking is particularly efficient, when the absorber is positioned such that (a) 2, (b) 3 or (c) 4 pulses collide here and cooperate in the opening of the shutter:



Another scheme for generating very short pulses uses traveling wave excitation in a transverse pumping geometry:



Even dyes with very low fluorescence quantum yield (on the order of 10^{-4}) can be used to generate ps-pulses with high efficiency.

V. Further Reading

Dye Lasers, F.P. Schäfer ed., 3rd edition, Springer 1990