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"TUNED DYE LASERS"

by

S. Chudzynski

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TUNED DYE LASERS

by

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Warsaw University

The first dye laser was built by Sorokin and Lankard in 1966. This launched the rapid development in lasers whose functional element is a solution (liquid or solid) of fluorescing organic dyes. The number of articles devoted to this problem amounts to a few hundred items at the moment. A vast amount of literature devoted to methods of obtaining laser action in organic substances and a list of "lasing" dyes can be found in the survey articles of Bass, Snavely, and others [1-7]. The enormous interest in organic compounds as laser materials is a result of the fact that they permit us to build tunable, continuous lasers (at present, in the 280 to 1175 nm range) and to obtain ultrashort light pulses (shorter than 3 ps) that are also tunable over a large region of the spectrum.

In the present article we shall discuss the physical principles of the dye laser as well as the practical methods for tuning it and its anticipated applications.

Laser Action in Solutions of Organic Dyes

The principle of the dye laser follows from the spectroscopic characteristics of dyes. We shall discuss them briefly. Since the molecules of a dye are systems numbering as many as a few hundred atoms, their vibration-rotation sublevels in the electron energy states create continuous bands in reality. Fig. 1 depicts these energy levels in a diagram. It is a typical one, although the distances between electron bands naturally depends on the

chemical compound. In thermal equilibrium at room temperature, only the lowest sublevels of S_0 singlet state are filled[1]. Optical pumping, accomplished with a flash lamp or laser (ruby laser, for example) will transfer the molecule from the S_0 state to the rotation-vibration sublevel of the S_1 singlet state, from where they will drop down to the lowest sublevels of the S_1 [Translator's Note: presumed to be S_1; subscript is not legible] state within 10^{-11} to 10^{-12} seconds (non-emitting transfer). From the S_1 level, the dye molecule can return to the S_0 state, emitting photons of less energy than the photons absorbed, and the fluorescence spectrum being is displaced toward the long wavelength region (Fig. 2). The lifetime τ is on the order of 10^{-9} seconds with respect to this transfer. Thus, thermalization of the S_1 state (i.e., a filling of the rotation-vibration sublevels within the S_1 state according to the Boltzmann distribution law for the given temperature) occurs before fluorescence appears.
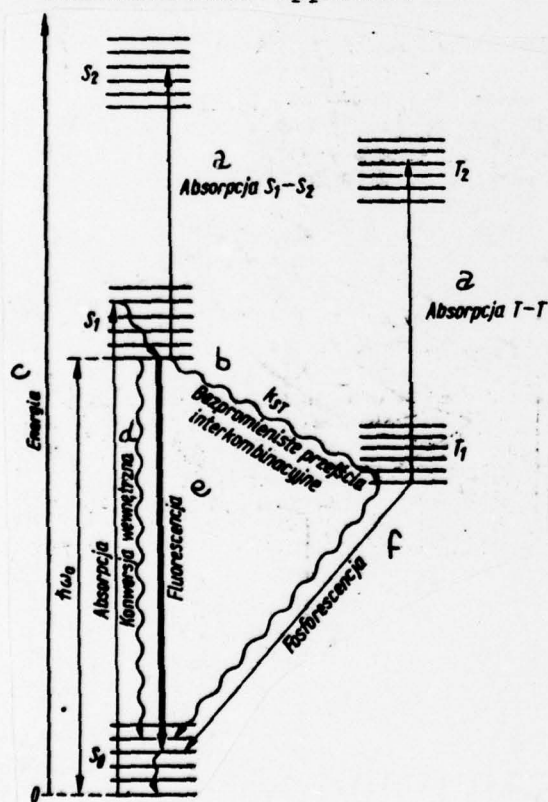

Fig. 1. Diagram of energy levels of dye molecules: wavy arrows denote non-emitting transfer, straight arrows denote optical transfer. Legend: a) Absorption; b) non-emitting inter-combination transfer; c) energy; d) internal conversion; e) fluorescence; f) phosphorescence

Fig. 2. A - cross section for absorption from the S_0 state, E - cross section for emission induced between the singlet states, T - cross section for absorption from the T_1 triplet state. Conditions of measurements, Φ: conc. 10^{-4} M; during measurement measurements of τ the concentrations amounted to 10^{-6} M [10]

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Excited molecules also ~~emit~~ release energy by means of non-emitting transfers - internal conversion, or non-emitting transfers between states with the same parity (S-S or T-T), or intercombination transfers between states of different parity (S-T). The probability of transfer k_{ST} for these latter ones is on the order of 10^3 to 10^8 s^{-1}. The ratio of the number of photons emitted from the S_1 state to the number of photons absorbed is called the quantum efficiency of fluorescence ϕ.

The lifetime τ_T of molecules in the T_1 state is on the order of 10^{-7} to 10^{-4} s (the metastable state). Hence, molecules return to the fundamental state either by emitting (phosphorescence) or without emitting. Molecules returning as a result of the previously mentioned transfers to higher sublevels of the S_0 state undergo thermalization. This process, however, is not always as fast as for the S_1 state. It follows from article [8] that the thermalization time for cryptocyanine in glycerol is on the order of 10^{-6} s. There is also absorption from the T_1 and S_1 states to the T_2 and S_2 states. It turns out that the absorption band (triplet-triplet) overlaps the fluorescence band (Fig. 2). Overlapping of the absorption band from the S_1 state on the fluorescence band is also possible. These are effects that impede laser action (self-absorption of laser radiation).

Laser action develops owing to transfers induced between the lowest sublevels of the S_1 state and the higher sublevels of the S_0 state (in the region of long wavelength fluorescence). Consequently, a dye laser is a four-level laser and the inversion of filled levels necessary to obtain laser action is not large. We reach the threshold of laser action at frequency [2] ω in a dye laser when the amplification of the photon beam N along the axis of the laser is at least equal to zero [2,5,9,10].

_____

[2] By the term "frequency" [Note: Polish term "czestosc" = rate or frequency] we shall understand in this article $\omega = \pi\nu$, where ν denotes frequency [czestotliwosc = frequency] - this is the nomenclature adopted among physicists, particularly in optics.

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$$G(\omega) = \frac{1}{N} \cdot \frac{dN}{dl} = \qquad (1)$$

$$= n^* \cdot \sigma^e_1(\omega) - m_0 \, \sigma^a_0(\omega) - n_T \, \sigma^a_T(\omega) - n^* \, \sigma^a_1(\omega) - r \geq 0$$

where:

n^*, n_0, n_T — concentration of molecules of dye in the S_1, S_0, and T_1 states;

$\sigma^e_1(\omega)$ — cross section (in cm^2) for absorption induced from the S_1 to the S_0 state at frequency ω;

$\sigma^a_1(\omega), \sigma^a_0(\omega), \sigma^a_T(\omega)$ — cross section (in cm^2) for absorption from the S_1, S_0 or T_1 state at frequency ω;

$r = -\frac{1}{2l} \ln(R_1 \cdot R_2 \cdot T_1 \cdot T_2)$ — describes the radiation losses that are not dependent on dye concentration;

l — length of the cuvette containing the dye solution;

R_1, R_2 — reflection coefficients of the cuvette windows;

T_1, T_2 — transmission of the laser mirrors.

The cross sections and fluorescence function (see below) introduced here are homologues of the Einstein coefficients B_{nm}, M_{mn}, and A_{nm}. Nonetheless, as McCumber [11] has shown, the relationships between them are different (for diffuse [broadened] electron states) than those from the generally known relations for narrow levels:

$$\sigma^a(\omega) = \sigma^e(\omega) \exp[\hbar(\omega - \omega_0)/kT]$$

$$\sigma^e(\omega) = f(\omega)\left[\frac{2\pi c}{\omega \cdot \eta(\omega)}\right]^2 \qquad (2)$$

where:

$f(\omega)$ - the fluorescence function normalized so that $\int f(\omega)d\omega = \Phi/\tau$,

$f(\omega)d\omega$ -   denotes the possibility of emission of a spon-
taneous photon having a frequency between ω,
and $\omega + d\omega$ in one second,

$\eta(\omega)$ -   refraction coefficient of the medium,

$h\omega_0$ -   distance between the lowest sublevels of the
S_0 and S_1 states.

Combining (1) and (2), we obtain:

$$G(\omega) = \{n^* - n_0 \exp[\hbar(\omega - \omega_0)/kT]\}\left[\frac{2\pi c}{\omega \cdot \eta(\omega)}\right]^2 \times$$
$$\times f(\omega) + n_T \sigma a_T(\omega) - n^* \sigma a_1(\omega) - r \geq 0 \qquad (3)$$

It is apparent from Eq. (3) that it is quite easy to achieve considerable amplification for a frequency that satisfies the condition $\omega - \omega_0 < 0$, which denotes operation of the laser in a four-level system. In general, laser action does not occur if

$$\sigma a_1(\omega) \geq \sigma e_1(\omega),$$

i.e., if the S_1 - S_2 absorption is too strong.

The laser material must be characterized by large values for the fluorescence function, i.e., a large quantum efficiency Φ, and a short lifetime τ of the S_1 state. It should be emphasized that in crystalline lasers (e.g., in a ruby laser) the upper laser level should be metastable and the luminescence band narrow. ~~Seeing that~~ Since the fluorescence function has a width of several tens of nanometers (Fig. 2), the conditions of generation are obtained for a wide frequency band. This is a fundamental characteristic and at the same time an advantage of dye lasers, which creates the possibility of tuning the laser by introducing a sel~~e~~ctive filter into the resonance chamber.

A very detailed analysis of Eq. (3) along with the equations that describe the evolution of filled levels is given in [1,2,3,9, 10,20]. It turns out that triplet-triplet absorption plays a major

role. For dyes whose probability of non-emitting intercombination
transfers is too large and for those that have a long lifetime in the T_1 state
($k_{ST} \cdot \tau_T$ is large), an interruption occurs in laser action due to
the accumulation of molecules in the triplet state. It is possible
to reduce the effect of this phenomenon by using additives that
quench the triplet state, e.g., oxygen, etc. [12,13], and by re-
moving "spent" liquid by rapid flow.

The effect of triplet absorption gives rise to the fact that,
although a few hundred dyes can be excited to laser action by
giant-pulse lasers, only about 50 can be excited using flash lamps -
in the majority of cases of special design, which ensures a short
pumping time on the order of 0.5 μs [7], and continuous laser action
has been obtained only in aqueous of alcoholic solutions of three
dyes: rhodamine 6G, rhodamine B and fluorescein and its derivatives.
The structural formulas of these dyes are given in Fig. 3 and
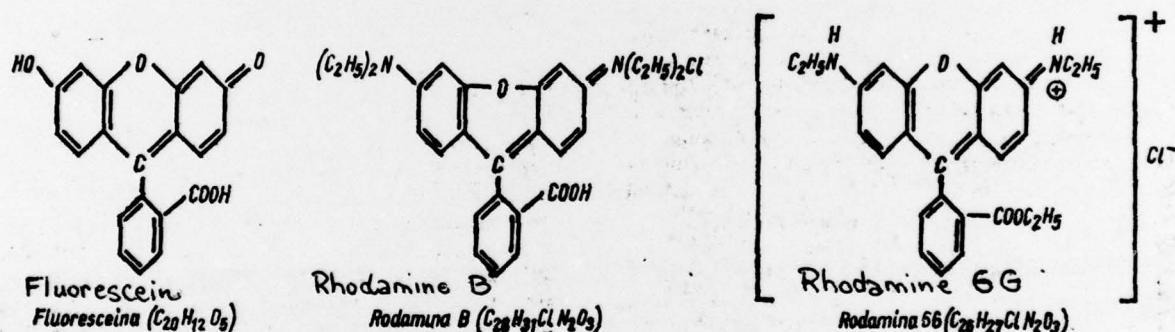their spectroscopic data are shown in the table.

Fig. 3. Structural formulas of three dyes: rhodamine 6G, rhodamine B,
and fluorescein

[Table]

Dye	Solution	Φ	l	$k_{ST} \cdot l_T$	l_T
Rhodamine 6G	ethanol	0.85	4.8 ns	0.9±0.2	0.1 μs
Rhodamine B	ethanol	0.6	3.1 ns		
Fluorescein (L)	0.1 n NaOH	0.85	4 ns		
Fluorescein (L)	water, pH = 9	1.0	4.5 ns		

L - data taken from Landolt Bornstein Zahlenwerte und Funktionen aus
Naturwissenschaften und Technik Neue Serie, Gruppe II, Band 3,
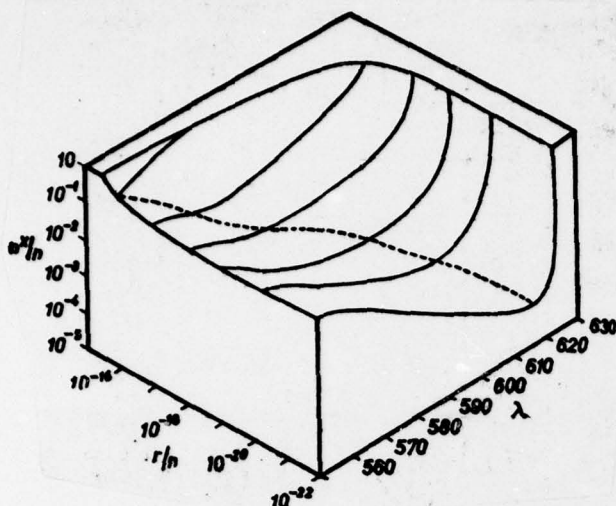Springer Verlag 1967, remaining data taken from [10]

Fig. 4. Relative filling of the excited singlet state at the threshold of laser action $(n^*/n)_{G=0}$ in rhodamine 6G at $k_{ST} \cdot \tau_{T=1}$ as a function of wavelength of laser action λ (nm) and normalized external losses r/n in cm^2/molecule of dye. The curves of constant losses (continuous lines) are the intersections of planes of constant losses with the surface of zero amplification. The broken line that connects the minima of these intersections denotes the threshold inversion of filled levels needed to obtain generation in a laser without a frequency selector; n - total concentration of dye particles [10]

Fig. 4 graphically depicts the solution to Eq. (3). In an actual experimental system we have specific losses, i.e., r/n (n is the concentration of dye). The minimum of the continuous curve defines for us the wavelength of laser action under threshold conditions along with the necessary threshold inversion of filled levels, ~~at the same time.~~ It is apparent that an increase in losses in the resonator will cause a rise in the threshold and a displacement of the wavelength of laser action in the direction of the short wavelengths. With losses of r/n = const., an increase in pumping (i.e., n*/n) will manifest itself by a broadening of laser action, mainly in the direction of the short wavelengths, particularly with a large Q factor for the resonance chamber [14]. In general, the band of laser action when a single dye is used depends on its concentration, the type of solvent, the length of the cuvette containing the liquid, the power of pumping and the temperature. Moreover, the frequency of laser radiation does not remain constant, but changes during pumping unless frequency selectors are used.

The direction of frequency changes at which the laser will begin to oscillate (with a given inversion of filled levels) with a change in the laser parameters: cuvette length l, concentration of dye n, transmission of the mirror T, is defined by the empirical euqation [15]

$$\frac{[\ln (T/l \cdot n)]}{d\omega} > 0 \qquad (4)$$

Tuning Dye Lasers

If we insert a tuned frequency selector into the resonance chamber of a dye laser that ensures a high Q factor for only one frequency, then it is possible to obtain laser action at almost any frequency in the entire fluorescence band. The systems used are shown in Fig. 5. In part a of this figure, tuning is accomplished by using a reflecting diffraction grating, ~~being both a~~ which at the same time is a single mirror of the laser resonator connected with a thin (about 1.0 mm) Fabry-Perot interferometer. As is evident, the grating is in the autocollimation position (Littrow). Gratings are used with a line density from 300 to 3000 lines per millimeter with slits profiled in such a way that ~~the~~ most of the light is deflected "backwards". Some gratings deflect up to 80% of the incident light in a single direction [16,17]. The angular dispersion δ and the free dispersion interval $\Delta\lambda$ are defined by the formulas (for a grating in the autocollimation arrangement):

$$\left. \begin{array}{l} \delta = \dfrac{d\lambda}{d\varphi} = \dfrac{\lambda}{2\,\mathrm{tg}\,\varphi} \cong 0{,}4 \ \dfrac{nm}{m\,rad} \\[4mm] \Delta\lambda = \dfrac{\lambda^2}{2a\,\sin\varphi} \simeq 60 \ nm \end{array} \right\} \tag{5}$$

if λ = 600 nm; the grating constant a = 0.5 μm (2000 lines/mm) and the angle of reflection ϕ^{φ} (angle of beam diffraction) is about 37°.

For the Fabry-Perot interferometer

$$\left. \begin{array}{l} \delta = \dfrac{d\lambda}{d\varphi} = \lambda\,\mathrm{tg}\,\varphi \\[4mm] \Delta\lambda = \dfrac{\lambda^2}{2t} \cong 0{,}18 \ nm \end{array} \right\} \tag{6}$$

The use of a grating and an interferometer offers the possibility of limiting the laser line below 10^{-3} nm [18]. Tuning of the laser is accomplished by rotating the grating and interferometer. Naturally, it is possible to use either just a grating or an interferometer, in which case broader lines are obtained.
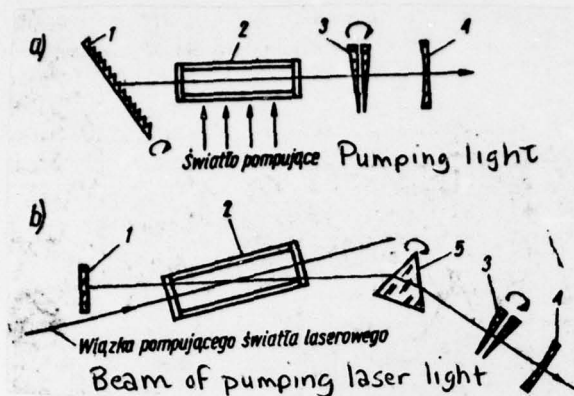
Fig. 5. Diagrams of tuned organic lasers: a) tuned using a diffraction grating; b) tuned using a prism: 1 - diffraction grating or dielectric mirror; 2 - cuvette containing the dye; 3 - Fabry-Perot interferometer; 4 - concave mirror; 5 - Brewster prism set up ~~under~~ *at* the angle of least aberration

In the system depicted in Fig. 5b a prism arranged ~~under~~ *at* the angle of least aberration is used as the preliminary selector. At the same time, the refraction angle is chosen so that for a single wavelength the laser beam strikes the wall of the prism ~~under~~ *at* the Brewster angle. In such an arrangement [19] the angular dispersion is expressed by the formula

$$\frac{d\lambda}{d\varphi} = \frac{1}{2}\left(\frac{d\eta}{d\lambda}\right)^{-1} \simeq 2,5 \text{ nm/mrad} \qquad (7)$$

since heavy phosphorus glass with a dispersion of 2.3×10^{-4} nm^{-1} and $\eta = 1.98$ for the D_1 sodium line was used. Although dispersion of the prism is worse than for the diffraction grating, the light losses in the prism are less than 1%, whereas in the grating they are about 50%. For precision a Fabry-Perot interferometer was also used in this case. Fig. 5 shows two different methods of dye laser pumping at the same time: perpendicular or oblique to the axis of the laser (in the last instance it is possible to pump merely using a second laser). Pumping along [parallel to] the axis of the laser is also possible.

Tuned Laser with Continuous Action

The functional elements in this laser are the solutions of dyes in water mentioned above, with a molar concentration on the order of 10^{-4} or somewhat higher with the addition of substances that

quench the triplet state. Fig. 6 depicts the optical system of
a laser from the Spectra company, model 70, pumped by an argon
laser line of 514.5 nm with a power in the range from 0.5 to 4 W.
Pumping occurs along the axis of the laser (the most efficient me-
thod among those mentioned above). It turns out that in order to
obtain laser action in the dye, it must be pumped with a power
density on the order of 100 kW/cm^2, which can be obtained only
when the exciting beam is collimated to a diameter on the order
of 6-10 microns. This imposes a requirement on the design of the
dye laser resonator. It must be designed so that the half-width
of the mode generated by the dye laser is also on the order of
6 microns and does not vary by more than a few percent during la-
ser operation. The resonator is long enough to contain a fre-
quency selector. A detailed design is given in [20]. It turns
out that with a resonator 30 cm long, it must be a three-element
type. We can see in Fig. 6 that the resonator comprises a flat
sapphire plate 2 with a spray-coated dielectric mirror with mini-
mal transmission of the generated lines and transmission reaching
100% for the 514.5 nm line; a lens 3 with a 5 mm focal length,
placed at the same distance from the sapphire plate and the mirror
6 with 6% transmission and a curvature of about 30 cm, separated
from the sapphire plate by a distance somewhat greater than 30 cm.
The plate and a short-focal-length lens determine the limitation of
the five-millimeter cuvette through which the the dye flows at a
rate of 1 liter/min. Lens 1 serves to collimate the pumping beam
in the cuvette region. The mode selector comprises a prism and a
Fabry-Perot interferometer. Prism 7 with a spray-coated filter
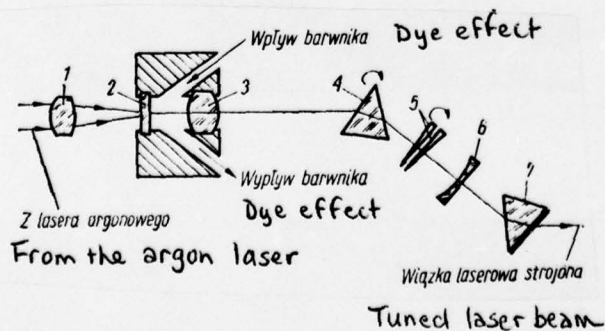serves to cut out the pumping line and direct the generated beam.

Fig. 6. Optical diagram of a
tuned organic laser with con-
tinuous action: 1 - focusing
lens for the 514.5 nm argon
laser beam; 2 - sapphire win-
dow with spray-coated dielec-
tric mirror; 3 - short-focus
lens; 4 - Brewster prism at
the angle of least aberration;
5 - Fabry-Perot interferometer;
6 - dielectric mirror, T = 7%;
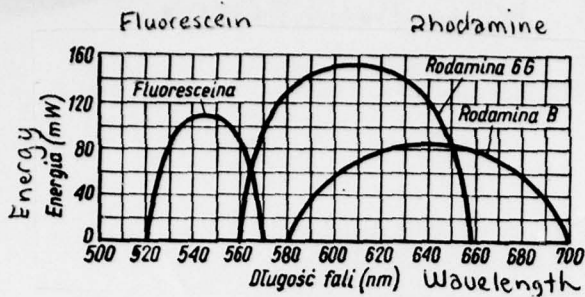7 - beam-orienting prism

Fig. 7. Spectral region of generation for the Spectra model 70 laser in Fig. 6

Fig. 7 depicts the region of the spectrum that this laser manages to cover. The parameters of this laser are of particular interest: The spectral width of the beam over the entire region of tuning is constant and amounts to 10^{-4} nm when a complete selector is used. The laser is equipped with a frequency control unit that permits selection of wavelength with an accuracy of 10^{-4} nm. The fluctuations in intensity are almost equal to the fluctuations of the pumping beam. The efficiency is about 10%. Fig. 8 provides an overall view of this laser.
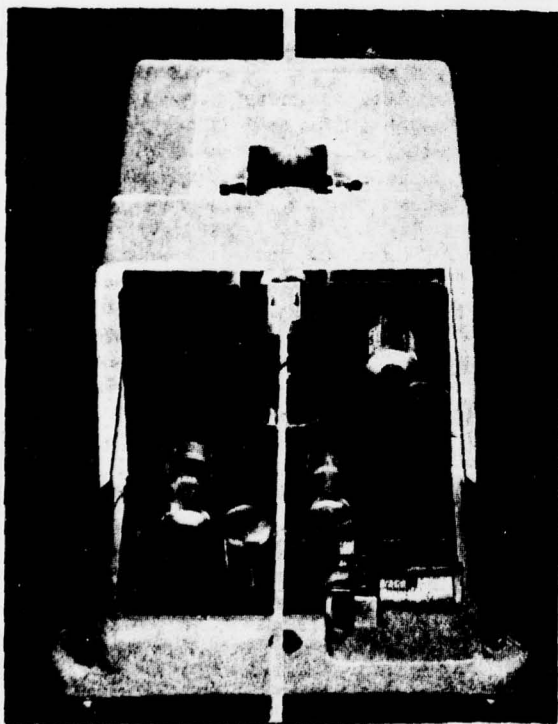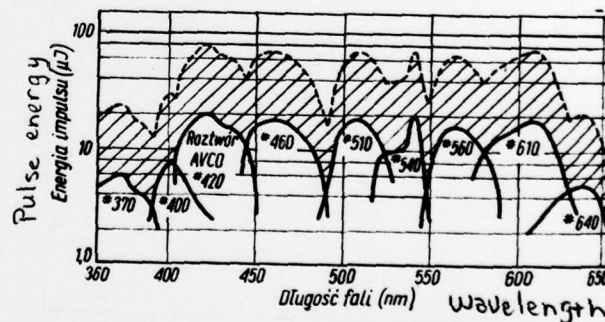

Fig. 8. Overall view of the Spectra model 70 dye laser

Fig. 9. Range of generation for the Avco tuned pulse dye laser

Pulse Laser Tuned in the 360-650 nm Range

If we abandon the condition of continuous laser operation, it is then possible to obtain laser action over a substantially wider

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spectral region with the aid of several dyes by using, for example, a pulsed nitrogen laser for pumping (λ = 3371 Å, flash time about 4 ns, power about 1 MW).

Fig. 9 shows the spectral region covered by a laser from the Avco company. In this instance, the chemical composition of the dyes were not given, but they are probably the ones mentioned previously; rhodamines, fluorescein and derivatives of another dye - coumarin [7, 17]. The energy of the flashes amounts to several μJ, the spectral width is 0.1 - 1 nm and the frequency of the flashes is up to 25 pulses/s.

Tuned Dye Laser Operating in the Mode Locking System

One of the most important properties of dyes is the possibility of obtaining laser action with mode locking in them. It turns out that for dyes, the pulses of light produced are extremely short, even less than 2 ps. The frequency spectrum of such a pulse, as follows from the Heisenberg principle, will not be narrower than $\Delta\nu$ =

$$= \frac{\Delta\omega}{2\pi} > \frac{1}{\Delta t} = \frac{1}{2}{}^{*}10^{12}s^{-1}$$

[* Note: this digit (or letter) is not clearly legible in original]

which corresponds to the wavelength interval

$\Delta\lambda = \Delta\nu\lambda^2/c \simeq 0.06$ nm for λ = 600 nm.

If, as in Fig. 5, we insert a chamber with a special organic compound that enables us to produce laser action of the mode locking type into the laser chamber in the appropriate place, then the frequency selector will limit the spectral width of the pulse to a value that derives from the uncertainty principle. Thus, we will be able, for example, by rotating the Fabry-Perot interferometer, to obtain

-12-

frequency tuning of the ultrashort light pulses. Arthurs, Bradley and Roddie [21,22] obtained such an effect for rhodamine 6G, rhodamine B and cresyl violet. These researchers managed to obtain picosecond light pulses, tuned in the 580-700 nm range.

Summary

Dye lasers are now becoming the most modern and the most promising family of lasers. The following should be included among the successes of this new technology: laser action over the entire visible spectrum including the near ultraviolet has been obtained, very simple and efficient frequency conversion has been achieved in the optical range (recently, 30% efficiency was obtained from a dye laser pumped by an argon laser), and picosecond light pulses with tuned wavelength have been produced. It should also be emphasized that a dye is inexpensive and generally available as a laser material. The following applications of dye lasers are possible:
- conventional spectroscopy, particularly microspectroscopy and nano- and picosecond spectroscopy,
- investigations of resonance phenomena (selective pumping of atoms or molecules),
- color holography,
- wideband light amplifiers (the amplification factor for certain dye solutions is expressed by a figure of 40 dB/cm),
- investigation of nonlinear phenomena caused by giant light pulses of different frequencies.

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Translation of non-English references:

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[5] "Generation of Complex Organic Compounds"

[14] "Rhodamine 6G Dye Laser", Fifth All-Poland Conference REK, 4/24-27/72

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