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UV DYE LASERS

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Introduction.

The most important property of visible dye lasers, that is continuous wave length tuning, stimulated the search for dyes^{x)} capable to lase in UV. And they were founded in 1968¹. Now the need in tunable UV lasers for applications in spectroscopy, photochemistry, isotope separation, remote air and sea probing, etc. is only more clearly seen. The object of this paper is to review shortly some recent advances in UV dye lasers.

^{x)} We use the term "dye" here only due to tradition. Active media of the lasers in question are certainly not dyes in common sense of the word.

Short Wave Length Limit of a Dye Laser Light.

The shortest wave length emission from organic compounds observed is the fluorescence of simple linear saturated hydrocarbons near $206-209 \text{ nm}^{2-4}$. It follows from the experiment²⁻⁴ and the theory⁵ as well that the efficiency of this fluorescence is very low. Branched and cyclic saturated hydrocarbons fluoresce at some longer wave lengths and have by an order of magnitude higher quantum yields. From published data for one of the best compounds of the latter type, bicyclohexil (fluorescence life time is 1,6 ns, quantum yield $\eta_{fl} = 0,02$, fluorescence band FWHM $\Delta\nu_{fl} = 8500 \text{ cm}^{-1}$, $\nu_{fl}^{\max} = 44200 \text{ cm}^{-1}$)^{4,6} one can estimate the stimulated emission cross section σ_{31} to be of the order of 10^{-20} cm^2 . Therefore it is necessary to have concentrations of excited molecules of about 10^{19} cm^{-3} in order to get an amplification factor of $0,1 \text{ cm}^{-1}$. To obtain these very high excited singlet state populations one needs a 4 to 5 orders of magnitude more powerful VUV pump light source than sources commonly used to pump dye lasers. There is no such sources for the time being. But even if they were, laser action in saturated hydrocarbons would be highly improbable because of inevitable efficient photochemical decomposition of molecules pumped by a powerful VUV radiation with photon energy close or equal to the ionization potential of the compounds in liquid state. In such conditions photochemical decomposition may be the main route for deactivation of molecular excited states.

For the same reason hydrocarbons with isolated multiple bonds do not seem to be promising. Organic compounds with a small conjugated π -electron systems and heteroatoms are much more perspective. Molecules of this kind rather strongly absorb radiation in the 200-280 nm range and fluoresce near 300 nm. Quantum yields can be as high as 0,67⁷. Sufficient pumping can be provided by the 4-th harmonic of the Nd³⁺-glass laser radiation or fast flashlamps. Thus the short wave length limit of dye laser radiation stands near 300 nm.

Laser Pumped UV Dye Lasers.

Since the first report on UV dye lasers¹ the wavelength of the p-terphenyl dye laser, 340 nm, remains the shortest one. This fact alone shows that it is not a simple matter to find a good compound to be capable for laser action in the spectral range between 340 and 300 nm.

We have made attempts to achieve laser action in a number of organic compounds which fluoresce between 290 and 340 nm. Among them there were benzimidazol, 2-methyl-benzimidazol, durene, anysol, indole, indazol as solutes in ethanol, water or cyclohexane. Quantum yield η_{fp} for these solutions varies from ~ 0,25 (durene in ethanol⁸) to 0,67 (benzimidazol in water, pH7⁷) and the estimated stimulated emission cross section σ_{31} varies from $0,1 \times 10^{-17} \text{ cm}^2$ (durene in ethanol) to $1,4 \times 10^{-17} \text{ cm}^2$ (indol in ethanol) as

compared to $\eta_{fl} = 0,93^8$ and $G_{31} = 15 \times 10^{-17} \text{ cm}^2$ for p-terphenyl in cyclohexane. Solutions were transversely (in some cases longitudinally) pumped by the 4-th harmonic of the Nd^{3+} -glass laser radiation with a pulse power of 1-4 MW. After pumping beam focusing by the cylindrical lens ($f = 160\text{mm}$) the maximum power density on the cell reached more than 100 MW/cm^2 . At such a high power densities the quartz cells broke after several pumping pulses and sometimes even after the single one. The cavities used were as short as possible ($\sim 10 \text{ mm}$) with flat high reflection dielectric or aluminized mirrors. But though the maximum intensities of pump light were about 1000 times that of the threshold value for p-terphenyl lasing at the same experimental conditions the generation threshold for compounds tested was never reached.

The fluorescence quantum yield is known often to be higher at low temperatures as compared to room ones due to slower intersystem crossing rate (for compounds with large $S_0 - S_1$ level separations)⁹. This leads to lower thresholds of low temperature generation 10^{-12} . We have carried out such experiments as well. But the generation thresholds in low temperatures glasses were not reached either.

Rather poor spectral properties of compounds can not be the only cause which prevents laser action in the solutions mentioned and alike. Evidently strong transient fluorescence and/or pump radiation absorption take place. Since an important influence of the last phenomenon on the generation properties has been pointed out ^{13,14}, its wide occurrence

has been found to take place ¹⁵⁻²⁰. Transient pump absorption not only causes higher generation threshold and worsens laser efficiency²¹, but also leads to the decomposition of an active compound due to stepwise two photon photochemical reactions. Even the best UV dye, p-terphenyl, which has the lowest induced pump ($\lambda_p = 265$ nm) absorption cross section among compounds studied ^{19,20}, suffers photoionization detected by transient photoconductivity in solutions ²². Therefore to get laser action in 340 to 300 nm spectral band one evidently must look for an organic compound with much better spectral and photochemical properties than those of compounds mentioned above.

So the full list of UV dye lasers known to us looks like that presented in table. Inspecting the table one must emphasize an important recent advance, namely vapour phase UV generation in POPOP. Firstly obtained ²³, then studied by Borisevitch and coworkers ^{24,25} and repeated by several groups of authors ²⁶⁻²⁸ vapour phase dye laser action is the real step towards a possible nonoptical dye laser pumping. The p-terphenyl seems to be another good candidate for UV vapour phase lasing.

The continuous tuning ranges indicated in table correspond to single solutions and selective cavities. To enlarge tuning range one can use binary solutions ¹³, which the sum amplification band is more wide than separate component bands. The simple theory ²⁹ which takes into account an inevitable in such a system intermolecular singlet-singlet energy transfer, satisfactorily explains experimental re-

Compound	Sol-	Laser wa-
	vent	ve length
		nm
p-terphenyl*	C	341
	C, E	330-362
2,5-diphenyl-1,3,4-oxadiazole (PPD)*	E	347
2,5-diphenyl-1,3-oxazole (PPO)	C	357
	T	365, 381
2-biphenyl-5-(4'-butylphenyl)-1,3,4-oxadiazole (butyl-PBD)	T	357-395
2,5-di-(4'-methoxyphenyl)-1,3,4-oxadiazole**	E	359, 372
	E	345-385
p-quaterphenyl	T	362-390
2-(4'-chlorophenyl)-5-phenyl-1,3-oxazole	C	363, 382
2-(4'-methoxyphenyl)-5-phenyl-1,3-oxazole	C	364, 383
2-(4'-methoxyphenyl)-5-phenyl-1,3,4-oxadiazole	E	365
	E	345-385
2-(4'-ethylphenyl)-5-phenyl-1,3-oxazole	T	366, 384
2,5-di-biphenyl-1,3,4-oxadiazole (BBD)	T	372-407
2-(4'-isopropylphenyl)-5-phenyl-1,3,4-oxadiazole	T	370
2-phenyl-5-biphenyl-1,3,4-oxadiazole (PBD)*	T	373
	T	385, 399
	T	366-398
2,5-diphenyl-furan (PPP)*	T	374
	T	367-380
2,5-di-(α -naphtyl)-1,3-oxazole (α -NNO)	C	374
β -naphtylen-oxide	E	375
2-(β -naphtyl)-5-phenyl-1,3-oxazole (β -NPO)	C	378
2-(α -naphtyl)-5-(β -naphtyl)-1,3,4-oxadiazole (α N β ND)	C	378
1,4-diphenyl-butadiene	T	383
2-biphenyl-5-phenyl-1,3-oxazole (BPO)	C	384
4-methylumbelliferone	E	385-457
2-(α -naphtyl)-5-phenyl-1,3,4-oxadiazole (α -NPD)	T	390
1,4-di-(benzoxazole)-benzene	Ch	390
2,5-di-(α -naphtyl)-1,3,4-oxadiazole (α -NND)*	T	391
	T	385-417
2-styryl-5-biphenyl-1,3,4-oxadiazole	T	391
2-styryl-5-(β -naphtyl)-1,3,4-oxadiazole sodium salicylate	T	392-430
amino-benzoic acid	E	395-418
2-(α -naphtyl)-5-styryl-1,3,4-oxadiazole	T	399
2-(α -naphtyl)-5-phenyl-1,3-oxazole (α -NPO)**	T	400

1) The solutions of compounds marked by an asterisk are active media of flashlamp pumped UV dye lasers as well (see^{49b,50-53}), (one of them marked by a double asterisk lasers in a binary solution with p-terphenyl⁵²). The record of the type 330-362 means the continuous tuning range and that of the type 365, 381 means two bands generation. C-cyclohexane, E-ethanol, T-toluene, Ch-chloroform.

sults²⁹. Depending on contour of fluorescence bands, their respective position and partial component concentrations one can obtain laser action in a binary solution at any wave length inside the sum amplification band in a nonselective cavity.

Tunable UV Light Generation by Methods of Non-Linear Optics.

Second-harmonic (SH) of and sum frequency with tunable dye laser light firstly reported in 1968³⁰⁻³² is now understood to be the only effective way to get tunable coherent UV radiation with wave length beyond 300 nm. While direct laser action seems to be probable in the 340 to 300 nm spectral range but yet no dye to lase in it found this same way is the only one to obtain tunable UV laser light with wave lengths shorter than ca. 330 nm (see table).

Tunable UV light as SH of a dye laser emission can now be obtained almost at any wave length from ca. 230 to 385 nm (or longer)^{30,33,34}, the short wave length limit being determined by phase-matching conditions in LFM crystall³³ (this limit for KDP and ADP is near 260 nm). SH power and efficiency of generation as high as 1 MW³⁵ and 18%³⁶ have been reported. Tunable cw SH have also been reported³⁷.

Phase-matching conditions for sum frequency generation (SFG) in widely used nonlinear KDP and ADP crystals (at room temperatures) permit one to get tunable radiation with short wave length limit of about 213 nm. Tunable SFG with wave lengths beyond the short wave length limit for SHG and

near the SFG limit have recently been obtained in the 216 - 226³⁸, 218 - 219³⁹ and 216 - 234 nm⁴⁰ ranges by summing Nd³⁺-glass laser fundamental and/or its harmonic frequencies with those of a dye laser in different schemes of three^{38,39} and four⁴⁰ frequency non-linear interaction. Sum frequencies in 241 - 327 nm range have also been reported^{32,39,41-44}, but SHG as more simple and effective is better than SFG for this spectral range (a new non-linear crystall, KB₂O₅, having phasematching condition for SHG down to 216,8 nm has recently been discovered⁶⁸).

Sum frequencies in VUV region can be obtained by frequency mixing in metal vapours⁴⁵. A method which ensures an increase in efficiency of this non-linear interaction by several orders of magnitude have recently been offered^{46,47} and the shortest wave lengths corresponded to the third harmonic of a dye laser near 155 nm and to the sum frequency tunable in the 177,8 - 181,7 nm range been obtained⁴⁶. The authors⁴⁶ have stated the short wave length limit in this method to be near 80 nm.

There is in principle at least one more way to get tunable coherent UV light. This way is to generate high order anti-Stokes stimulated Raman scattering (SRS) in an ordinary Raman medium pumped by a powerful tunable dye laser⁴⁸ or low order Stokes SRS in a various Raman media pumped by a powerful UV laser lines. But mainly because of SRS having threshold and some other evident shortcomings not to be discussed here both these approaches do not seem promising.

Flashlamp Pumped UV Dye Lasers.

Laser pumped UV dye lasers have low both the overall efficiency and the output pulse energy. By use of flashlamp pumping these parameters of an UV dye laser can be considerably increased. Therefore much attention has been paid to development of the fast flashlamps and low inductance discharge circuits capable to provide light powers sufficient to pump UV dyes to lasing thresholds ⁴⁹⁻⁵³. All systems developed ⁴⁹⁻⁵² except the recent one ⁵³ to be discussed later are similar in essential details. They consisted of a short (35 - 50 mm) coaxial xenon filled flashlamp with a current return lead, a high voltage low inductance capacitor of up to 0.08 μ F and a switch connected in series to the lamp. The coaxial lamp discharge annulus varies from 0.3 to 0.5 mm. For such a discharge circuit the light pulse rise time of 50 to 80 ns, its FWHM pulse time of 150 to 200 ns, and the maximum electrical energy of about 20 J are typical values. Up to date the efficiency of flashlamp pumped UV dye laser (they are marked by an asterisk in table) are of the order of 0,01%.

The system described in a recent paper by Morrow and Price ⁵³ considerably differs from above mentioned ones. The much larger lamp used differs in both the construction and all demensions except the discharge annulus (0,5 mm). Being fully fused it has no epoxy sealed joints. By-electrode ballast volumes help it to stand large dynamic stress-

es and to dissipate 100 J electrical energy deposited into discharge without breaking. The lamp light pulse rise time and p-terphenyl dye laser pulse FWHM are much longer and equal to 150 and 500 ns respectively. Comparatively high output laser pulse energy of 0,01 J was obtained.

In general less dyes show laser action under fast flashlamp pumping as compared with laser pumping and some dyes showing good performance in the latter case have high generation thresholds or do not lase at all in the former one ^{49b}. There can be several causes leading to high laser thresholds in case of flashlamp pumping. Most of them are common for both laser and flashlamp pumping but their influence on generation properties of a dye laser is much more prominent in the latter case. The well known triplet state effects leading to excessive depletion of the dye ground state population or to laser emission absorption by triplet molecules are maybe the simplest harmful effects in a flashlamp pumped dye laser. Due to fast flashlamp pump emission being that of a high temperature (ca. 40000-70000K) black-body radiator with the continuous emission spectrum transient pump absorption by excited singlet and triplet molecules necessarily takes place resulting in considerable pump energy losses, higher thresholds, more prominent thermal effects, dye photolysis etc. There is also one peculiar important phonomenon inherent only to flashlamp pumping which has been pointed out by Stepanov and Batyrev ⁶⁹. Namely this is the amplification of pump radiation by stimulated emission in the fluorescence band of an active medium.

This phenomenon is in fact an additional effective route for deactivation of excited singlet states with obvious undesirable consequences.

For UV dye lasers pumped by short flashlamp pulses the thermo- and acousto-optical cavity distortions are less important in determining output laser parameters than for long pulse flashlamp pumped visible dye lasers. As pointed out by Smolskaya and Rubinov ⁵⁴, the cavity thermo-optical distortion at short pulse pumping should be described by the refractive index derivative value $(dn/dT)_v$ (at constant volume) rather than $(dn/dT)_p$ (at constant pressure) commonly used. The former value is much less than the latter one and can even have the inverse sign ⁵⁵. By the end of a dye laser pulse the shock wave runs only a small distance into the bulk of solution so that it also does not considerably disturb its optical homogeneity.

Conclusion.

The further search of the UV dyes is necessary not only to fill the gap between expected and already realized wavelength limits (300 - 330 nm) but also to find good dyes capable to lase with low thresholds primarily under flashlamp pumping. It is also necessary to further study photophysical and photochemical processes which take places in solutions under powerful photoexcitation to fully understand loss sources and to find ways to lessen their influence on a dye

laser performance and long term photostability of active media. From practical point of view flashlamp pumped UV dye lasers are of particular importance and further work are needed to improve considerably their output parameters and also to run them in quasicontinuous high pulse repetition rate mode of operation.

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