A tutorial on the technical analysis of dye lasers and short review of their novel applications

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Abstract

The field of Dye lasers has recently started evolving briskly, and many researchers have carried out serious investigations for exploring the newer applications of these lasers. This paper gives the technical analysis of the dye lasers, besides presenting a short review of their recent important novel applications. It is expected that the paper should be useful for the new entrants in the field, and also the researchers engaged in exploring the newer applications of dye lasers.

Keywords: Laser Dyes, Energy States of an Excimer, Novel Applications of Dye Lasers.

Resumen

El campo de los láseres de colorante ha comenzado recientemente a evolucionar rápidamente, y muchos investigadores han llevado a cabo investigaciones serias para la exploración de nuevas aplicaciones de estos láseres. En este trabajo se da un análisis técnico de los láseres de colorante, además de presentar una breve reseña de sus novedosas recientes aplicaciones importantes. Se espera que el documento sea útil para los principiantes en este campo, así como a los investigadores que trabajan en la exploración de nuevas aplicaciones de los láseres de colorante.

Palabras clave: Láseres de colorantes, Estados Energía de un Excimer, Aplicaciones Novedosas de Láser de colorante.

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I. INTRODUCTION

A dye laser is a laser based on using an organic dye as the lasing medium, mostly in the form of a liquid solution, and has the advantage as compared to the solid and gaseous lasing media mainly in the sense that a dye can usually be used for a much wider range of wavelengths, and this aspect of the wide bandwidth makes them useful as tunable lasers (tuning being done by mounting a prism or diffraction grating in the path of the beam), in addition to the fact that the dye can be replaced by another type of dye for generating different wavelengths with the same laser, with slight replacement of the optical components. It is important to note that in certain cases, solid state dyes like dye doped organic matrices are used as the gain medium, and these lasers are called as solid state dye lasers (SSDLs). The work on dye lasers started in the mid 1960s [1], and some papers were also published in 1970s – 1980s [2, 3, 4]. Valdmanis et al [4] have described an ultrashort pulse laser, which under specific operating conditions, balances the mechanisms of conventional passive mode locking and soliton like pulse shaping in a single resonator to generate 27 fsec optical pulses emitted directly from the laser. After that, some regular progress has been observed in their development, with great efforts being made in the last decade on the evolution of the subject.

A dye laser is fabricated by mixing an organic dye with a solvent, and then either circulating it through a dye cell (or cuvette), or streaming through the air by using a dye jet (to avoid the reflection losses). Just like the conventional lasers, its fabrication requires some basic components like: (i) a high energy source of light is required for pumping the liquid beyond its lasing threshold, (ii) a fast discharge flashlamp (or an external laser) , (iii) a set of mirrors (one fully reflecting ~ 99.99% reflectivity, and the other , the output mirror with 80-85% reflectivity) to oscillate the light produced by the dye's fluorescence, amplified with each pass through the liquid. Since the liquid dyes have very high gain as laser media, the beam requires making a few passes through the liquid for achieving the full design power, and thus resulting in the high transmittance of the output coupler. Care is taken for coating or anodizing the pump cavities or making them of a material that does not reflect at the lasing wavelength when being reflected at the pump wavelength. The schematic of a dye laser has been shown below:

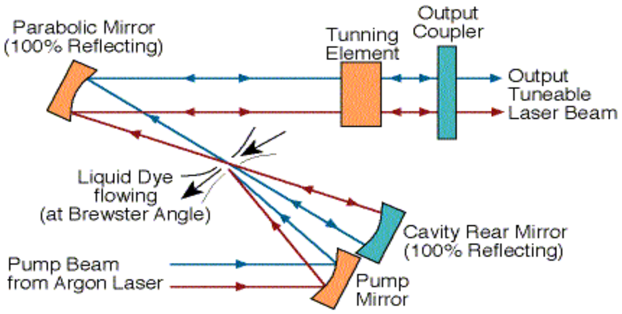

FIGURE 1. Schematic of a Dye Laser. Figure courtesy stwww.weizmann.ac.il.

The dyes used in these lasers have large organic molecules, which fluoresce, since the dye molecules are excited by the incoming light to emit stimulated radiation in the singlet state, in which the molecules emit light via fluorescence, the dye chosen being transparent to the lasing wavelength. Within a microsecond, or less, the molecules change to their triplet state very fast, in ~ a microsecond, in which state, light is emitted by phosphorescence, and in the process, the molecules completely absorb the lasing wavelength, and change the transparent dye into opaque. Great care has to be taken that the speed of circulation is very high, which helps in avoiding triplet absorption and also decreasing the degradation of the dye. The organic dyes have a tendency of decomposing by light, and that is precisely the reason that the dye solution is circulated from a large reservoir, through a cuvette or a dye jet, which avoids reflection losses from the glass surfaces and contamination of the walls of the cuvette, though for these advantages, alignment becomes more complicated. The frequently employed kind of dye laser uses a thin dye jet as the gain medium, in which case the dye molecules are used only for a short time within the pump and laser beam, and also have a long time for recovering before being used again. Time for recovery is required because of the tendency of organic dye molecules to become trapped in triplet states, and so ceasing to participate in the lasing process. In some cases, the triplet concentration is lowered by adding a triplet quenching agent to the dye solution. In some cases, the dye is pumped through a thin cuvette of some material like quartz, and the dye is enclosed in some transparent material, which has the advantage of easily obtaining a steady flow. Quartz is chosen because it is resistant to the laser light and the pump light, and the cuvette surfaces must have a high optical quality. The passively mode locked dye lasers yield very short pulses ~ few hundred fs. It is important to note that they are limited by our ability to saturate the absorber. The schematic of the passively Mode-locked Dye Laser has been shown below:

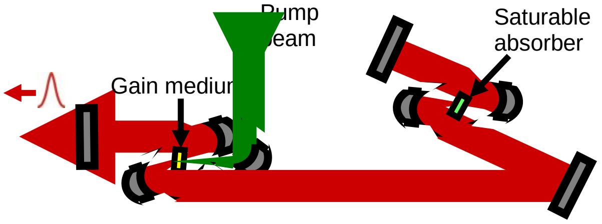

FIGURE 2 Mode-locked Dye Laser. Figure courtesy www.powershow.com/.

The fact that the liquid medium of a dye laser can be employed in any shape, leads to the possibility of using many different configurations. The most commonly employed configuration is the Fabry Pérot (FP) laser cavity, which is mainly used for flashlamp pumped lasers. The F.P. cavity consists of two mirrors, flat or curved, mounted parallel to each other with the laser medium in between them. In general, the dye cell is side pumped, having one or more flashlamps positioned parallel to the dye cell in a reflector cavity, which is mostly water cooled, to prevent the possibility of the thermal shock in the dye caused by the large amounts of near IR radiation produced by the flashlamp. The other common geometry is that of the axial pumped lasers, having a hollow, annular shaped flashlamp surrounding the dye cell, which has lower inductance to provide a shorter flash, and also the improved transfer efficiency. In some cases, we have coaxial pumped lasers having an annular dye cell, surrounding the flash lamp, which provides even better transfer efficiency, though with a lower gain due to the diffraction losses. It is a common practice to choose a ring laser design for continuous wave (CW) operation, in which the mirrors are positioned so that the beam follows a circular path. The dye requires to be pumped, which is normally done by an external laser like excimer laser, or frequency doubled Nd:YAG laser. The ring laser design behaves differently than the FP cavitiy, in that it does not generate standing waves, causing a phenomenon in which the energy is trapped in unused portions of the medium between the crests of the wave; and leads to the spatial hole burning, which results in a better gain from the lasing medium, and hence a better efficiency. In general, the dye lasers have some additional components like several frequency selective elements *e.g.* a birefringent tuner or a diffraction grating in Littrow configuration, for the wavelength tuning in a range of tens of nanometers. For the case of the narrow linewidth dye lasers, use is made of some more frequency filtering components like etalons, and also the sophisticated computer controlled tuning mechanics for precise output of the desired wavelengths. However, in case of the mode-locked lasers, with large emission bandwidth, use of a course wavelength control is adequate. It is interesting to note that these tuning elements provide a stable linear polarization of the output.

Thus we see that dye lasers are liquid lasers, having organic dyes solved in organic solvents, pumped by a laser, to emit light via fluoresence. The dyes are generally organic polyatomic molecules with conjugated π-chains, like - rhodamine, tetracene, coumarine, and stilbene; and the comon solvents are - methanol, ethanol, water or ethylene glycol. The additional chemicals are added , which prevent intersystem crossing and also prohibit degration of the dye. Some of the polyatomic organic molecules contain the conjugated double bonds. The electrons move freely within the whole chain, and may be described as a free electron in 1D potential well.

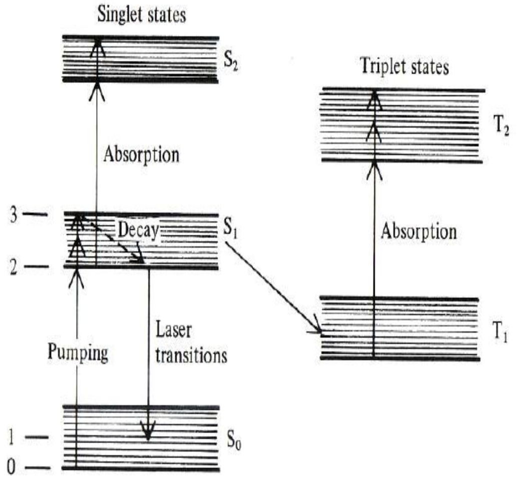

FIGURE 3. The Singlet and Triplet states of Dye Laser. Figure courtesy www.star.le.ac.uk.

There are three singlet states – S0, S1 and S3, and two triplet states T1 and T2. The pumping takes S0 to S1. The singlet S1 consists of two levels (2 and 3), and the singlet S0 consists of two levels (0 and 1). The vibrational and rotational levels remain unresolved in the liquids. The selection rules are: (i) $\Delta S = 0$, and (ii) S1 to S0 is allowed. These systems contain an organic dye in a solvent. Such dyes can be excited by absorption of short wavelengths and fluoresce by emitting at longer wavelengths. There are a large number of electronic energy levels in bands, resulting in a large number of possible LASER transitions, and hence these lasers are tunable.

Pumping is done optically using radiation from another laser, for example the Ar ion laser. The Fluorescence emission takes place from S1 to S0. There are three types of losses – (i) The inntersystem crossing – S1 toT1; (ii) The phosphoresence – T1 to S0; and (iii) Absorption – S1 to S2, and T1 to T2. There are certain problems faced while making a dye laser - (i) Short lifetime of the S1 state, (ii) Intersystem crossing and long lifetime of T1, and (iii) Thermal gradients produce refractive gradient.

II OPERATION OF DYE LASER

The operation is mainly based on the following points: There is a Pulsed laser action, with Circulation of dye solution, and pumping through another laser like - Nitrogen laser (UV-visible), Excimer laser (UV-visible), and Nd:YAG laser (visible). Since the excimer laser is very commonly empoyed for pumping, it is briefly disussed technically as given below:

The Excimer LASER is the Electron pumped LASER, based on the Dimer (excimer)/complex (exciplex) formation, and providing the LASER radiation: as a resut of the relaxation from excited state dimer to ground state as:
$e^- + A \rightarrow A^*$, and $A^* + B \rightarrow AB^* \rightarrow AB + h\nu$, where hν is the energy. After this action, immediately, disassociation of AB takes place as: $AB \rightarrow A + B$.

There are two important facts, that have to be noted, and these are: (i) The lower state does not exist, and (ii) There are no rotational/vibrational bands. The Energy states of an excimer are shown in the following figure:

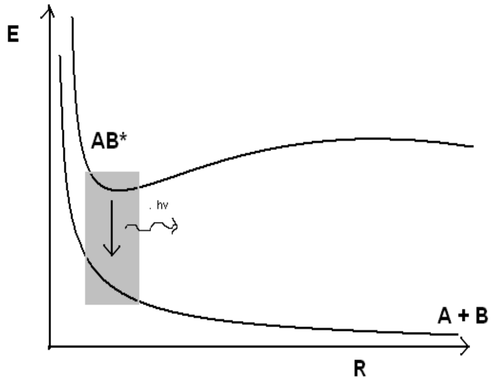

FIGURE 4. Energy States of an Excimer.

The excited dimmers are of the form - F2, Xe2, and the excited complexes (exciplex) are formed by the combination of rare gas (Ar, Kr, and Xe) atoms and halogen (F, Cl, and Br) atoms. The action can give various wavelengths depending upon the excited dimer. The repetition rate observed varies from 0.05 Hz to 20 kHz, and the laser beam is observed to be of high power ~ several 10 – 200W. Some of the Excimer gases and the related wavelengths emitted by them are given below:

Ar2 - 126 nm, Kr2 - 146 nm, F2 - 157 nm, Xe2 - 172 nm and 175 nm, ArF - 193 nm, KrF - 248 nm, Cl2 - 259 nm, N2 - 337 nm, and XeF - 351 nm.

III APPLICATIONS OF DYE LASERS

Dye lasers are really quite versatile in the sense, that in addition to their recognized wavelength, these lasers offer very large pulsed energies or very high average powers; e.g. the Flashlamp-pumped dye lasers are able to yield hundreds of Joules per pulse. Dye lasers are used in many applications including astronomy, as laser guide stars, medicine, and spectroscopy.

In the field of medicine, these lasers are applied on several areas, including dermatology, in which they are used to skin tone more even. Vascular skin lesions are known to contain oxygenated haemoglobin, which has the characteristic of strongly absorbing the visible light at 418nm, 542nm and 577 nm, and the pigmented skin lesions contain melanin, which has a wide range of absorption in the visible and IR wavelength regions. The difficulty in using the IR lasers is that they are very destructive because of the fact that they are absorbed by water in and around the skin cells. However, the aim of the doctor is to destroy the target cells and also at the same time not to harm the surrounding tissues, and this is achieved by using the short pulses, and thus reducing the heating of the damaged cells, and finally reducing the thermal injury which could result in scarring. In this treatment, help is also taken from the automated scanners, which reduce the chances of the overlapping of the treatment areas.

Many types of lasers including the argon, APTD, KTP, krypton, copper vapour, copper bromide, pulsed dye lasers and Nd:YAG. Argon (CW) have been used successfully to treat various types of vascular lesions, including superficial vascular malformations like the facial telangiectases, and poikiloderma of Civatte. However, the pulsed dye lasers have been found to be the most suitable for these vascular lesions because they are clinically very efficient and quite low risk profile. The dye lasers are found to be very effective because of two factors: (i) the wide range of possible wavelengths matching very close to the absorption lines of the certain tissues (melanin or hemoglobin), and (ii) the narrow bandwidth obtainable helping in reducing the possibility of damage to the surrounding tissue. Because of the same reasons, they are also used for various other applications like – treatment of port-wine stains and other blood vessel disorders, scars and kidney stones.

Dye lasers are very useful for carrying out the research work in spectroscopy, as they can be used to study the absorption and emission spectra of various materials, very efficiently due to many characteristics:(i) Their tunability, from the near IR to the near UV, narrow bandwidth, and high intensity allowing a much greater diversity than other light sources. These lasers are also used for the environmental pollution monitoring.

IV RECENT NOVEL APPLICATIONS AND CONCLUDING REMARKS

The field of dye lasers has drawn the increased attention of the researchers in the last decade. This is also because of the fact that apart from other uses of the lasers, their biomedical applications, especially for skin diseases, are making them specially very important. Some of the recent important novel applications have been reviewed here. Bornemann et al [5] have reported the first realization of a cw solid-state dye laser., in which the laser medium consists of a laser dye (Rhodamine 6G) dissolved in a photopolymer, and the UV cured solution is sandwiched between two Digital Versatile Disc (DVD) substrates, the resonator design being derived from a conventional liquid solvent dye laser geometry. It has been shown that the laser radiation can be tuned from 565 to 615 nm by using a birefringent filter, and a pump power of 2 W leads to a cw output power of more than 20 mW. Won [6] has discussed that the Optofluidic dye lasers, although attractive as miniature coherent light sources for integrated optics, have a ilimitation in that they require a solvent for the preparation of their liquid dye solution.

Choi *et al.* [7] have reported on the demonstration of liquid organic dye lasers free from solvent, based on 9-(2-ethylhexyl)carbazole (EHCz), so-called liquid carbazole, doped with green- and red-emitting laser dyes; and also have prepared both waveguide and Fabry-Perot type microcavity fluidic organic dye lasers by capillary action under solvent-free conditions. This work was accomplished by employing Cascade Förster-type energy transfer processes from liquid carbazole to laser dyes, for achieveing color-variable amplified spontaneous emission and lasing. It has been emphasized that this study provides the first step towards the development of solvent free fluidic organic semiconducting lasers and also demonstrates a new kind of optoelectronic application for liquid organic semiconductors.

It is now accepted that the Lab-on-a-chip systems made of polymers are promising for the integration of active optical elements, enabling *e.g.* on-chip excitation of fluorescent markers or spectroscopy. Wienhold *et al.* [8] have presented the diffusion operation of tunable optofluidic dye lasers in a polymer foil, and have demonstrated that these first order distributed feedback lasers can be operated for more than 90 min at a pulse repetition rate of 2 Hz without fluidic pumping. It has been reported that the Ultra-high output pulse energies of more than 10 μJ and laser thresholds of 2 μJ have been achieved for resonator lengths of 3 mm. Tunability of laser output wavelengths over a spectral range of 24 nm on a single chip has been accomplished by varying the laser grating period in steps of 2 nm. It has been emphasized that these on-chip lasers are suitable for a wide range of lab-on-a-chip applications, e.g. excitation of fluorescent markers, and surface enhanced Raman spectroscopy (SERS).

Rawat *et al.* [9] have designed and fabricated a dye cell to facilitate high repetition rate single longitudinal mode (SLM) operation with low viscosity solvents such as ethanol, and also have been able to eliminate the flow circulation (vortex) in the dye cell by reducing the flow cross section from 10 to 5 mm^2 with optimized flow entry. Flow visualization of various geometries in the dye cell has been carried out using commercial computational fluid dynamics (CFD) software, and it has been reported that the slit as well as tubular entry to the dye cell of cross section 1×10 mm^2 shows flow circulation (a vortex) near the entry to the dye cell. It has been highlighted that the time averaged SLM line widths of 400 and 175 MHz were obtained with a copper vapor laser (CVL) and Nd:YAG laser, respectively; and a single pulse line width of 315 MHz was obtained with a CVL pumped dye laser. It should be noted that these line widths are quite significant from some specific aopplications pont of view.

Klinkhammer *et al*. [10] have reported the fabrication and characterization of continuously tunable, solution processed distributed feedback (DFB) lasers in the visible regime, in which the continuous thin film thickness gradients have been achieved by means of horizontal dipping of several conjugated polymer and blended small molecule solutions on cm scale surface gratings of different periods. In addition, the optically pumped continuously tunable laser emissions of 13 nm in the blue, 16 nm in the green and 19 nm in the red spectral region have been obtained on a single chip. Passeron et al [11] have shown a treatment to prevent, by a large extent, the relapse of melasma. In the medical field, it is well known that the Melanocytes express vascular endothelial growth factor (VEGF) receptors 1 and 2 and neuropilin, and the VEGF and skin vascularization might play a role in the pigmentation processes, and therefore in melasma. It has been emphasized that by targeting the vascular component in melasma lesions, the used dye laser may decrease the melanocyte stimulation and subsequently the relapses. It has also been suggested that additional studies are required to confirm this result of the prevention of relapses, and also to optimize the treatment.

Gerosa *et al*. [12] have demonstrated an all-fiber dye laser, in which the dye solution is kept under flow, allowing for high repetition rate pumping. It has been reported that the threshold average pump power of 2.15 mW and conversion slope efficiency of ~8.5% have been achieved. Yang et al [13] have reported a highly sensitive stress probe based on pyrromethene 597 (PM597) doped elastic polydimethylsiloxane films. By sandwiching the dye doped elastic film with two plano dichromatic mirrors, a solid-sate microcavity laser with low laser threshold (0.2μJ) has been presented as a simple probing method for mechanical stress, which is monitored by the laser output spectra, and have demonstrated a resolution limit higher than 0.01 MPa. It is also interesting to note that they have achieved the photostability of PM597 doped into the microcavity laser higher than 7222 GJ/mol, and also have observed fast self-recovery on the laser output in less than 1 h, which is considered to be due to the diffusion of dye molecules. Song and Psaltis [14] have presented a tunable optofluidic dye laser with integrated elastomeric air-gap etalon controlled by air pressure, by fabricating the chip with polydimethylsiloxane (PDMS) via replica molding, which comprises a liquid waveguide and microscale air-gap mirrors providing the feedback. The system is based on choosing the lasing wavelength by the interference between two parallel PDMS-air interfaces inside the internal tunable air-gap etalon, and realizing the pneumatic tuning by inflating the air-gap etalon with compressed air. The results like - (i) a pumping threshold of 1.6μJ/pulse, (ii) a lasing linewidth of 3 nm, and (iii) a tuning range of 14 nm, have been achieved.

Rosacea is a commonly noticed chronic inflammatory condition, which is characterised by erythema, telangiectasias, papules, and pustules. There are many quite effective treatmentmechanisms for the papulopustular type. Laser therapy is found to be the most effective technique for the treatment of erythematotelangiectatic rosacea. Kashlan *et al*. [15] have proposed a novel technique for enhancing the response of rosacea patients being treated for erythema with pulsed dye laser, and have shown that pre treatment with forced heated air prior to pulsed dye laser leads to a greater response in rosacea patients with erythema and flushing. Thus, it is observed that novel applications of the dye lasers have been established by the researchers, on the basis of which, it can be concluded that the field of dye lasers applications is evolving at a brisk pace.

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