ABSTRACT

Dye lasers are the most versatile and one of the most successful laser sources known today due to their significant contribution to science and technology. Solid state dye lasers provide an attractive alternative to the conventional liquid solutions with the obvious advantages such as compactness, manageability, lack of toxicity or flammability and suppression of flow fluctuations and solvent evaporation, which would facilitate miniaturization and design of optical systems.

Most of the recent works have been done using either polymers or silica gels as the host media. High-damage-threshold polymer materials have played a significant role in the advancement of solid-state dye laser systems. Most frequently used polymeric material is the Polymethylmethacrylate (PMMA) because of its best optical transparency and high laser damage resistance. The main concern in the recent research has been the search for particular dyes and solid host that provide high laser damage threshold and long lifetime against photodegradation. Further improvements in the solid state dye laser require greater understanding of the photophysical properties of the dye molecules in polymeric matrices.

This thesis presents the spectral and nonlinear studies of organic dyes in liquid and polymeric environment. Dyes chosen for our studies are i)
Methyl Violet 2B, ii) Basic Fuchsins, iii) Fuchsins Acid, iv) New Fuchsins, v) Aniline Blue and vi) Cresol Red. PMMA modified with low molecular weight additives. Dye lasers are the most versatile and one of the most successful laser sources known today due to their significant contribution to science and technology. Solid state dye lasers provide an attractive alternative to the conventional liquid solutions with the obvious advantages such as compactness, manageability, lack of toxicity or flammability and suppression of flow fluctuations and solvent evaporation, which would facilitate miniaturization and design of optical systems.

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Aniline Blue and vi) Cresol Red. PMMA modified with low molecular weight additives (Ethanol and n-Butanol) is used as the host medium for these dyes. Material characterization of these dye doped polymers was done by Vickers hardness, FTIR, TGA and DSC measurements. Further absorption spectra, fluorescence spectra, and fluorescence lifetime measurements are also made to provide additional data for understanding the laser behavior.

Nonlinear characterizations are done using z-scan technique for opto-electronic applications like optical data storage and optical limiting. The z-scan technique is based on the intensity dependence of the nonlinear refractive index, a third order nonlinear optical phenomenon. The majority of the nonlinear optical phenomena are considered to occur in isotropic materials. The second order nonlinear effects being absent in isotropic medium, the lowest order of nonlinearity exhibited by these materials is of the third order. The basic idea behind z-scan is self-focusing. Self focusing occurs when light beam of nonlinear intensity passes through a medium. The refractive index maps the intensity pattern in the transverse plane.

The spectral behavior of the individual dyes in solid (MPMMA) and in liquid media (MMA, solvent, MMA+solvent) are studied. The spectral behavior of the dyes depends on the absorption and fluorescence of the dyes in the respective media.
The absorption and fluorescence spectral profiles of the dyes, in liquid and solid media, were identical. This indicates that at low concentration, these dyes exist in its monomeric form and that at such low concentrations, there is a given amount of free volume within the polymer matrix in which the dye molecules distribute themselves within it in a way similar to what happens in the dilute solution.

The nonlinear characteristics of the individual dyes in solid (MPMMA) and liquid media are studied. The third order nonlinear optical parameters of the dyes in liquid (dye in solvent) and in solid (dye doped polymer film) media were determined by using the Z-scan technique developed. The Z-scan technique is a simple but very accurate method to determine both nonlinear index of refraction $n_2$ and nonlinear absorption coefficient, $\beta$. Nonlinear index of refraction is proportional to the real part of the third-order susceptibility $[\text{Re}\chi^{(3)}]$ and the nonlinear absorption coefficient is proportional to $[\text{Im}\chi^{(3)}]$.

All the dyes showed a negative nonlinearity, which is observed from the Z-scan trace itself. The traces show a pre-focal transmittance maximum (peak) followed by a post-focal transmittance maximum (valley), indicating the dyes have negative nonlinearity. It is observed that the nonlinear absorption all the triarylmethane dyes can be attributed to a saturation absorption process.
The defocusing effect is attributed to a thermal nonlinearity resulting from absorption of radiation at the excitation wavelength. Localized absorption of a tightly focused beam propagating through an absorbing dye medium produces a spatial distribution of temperature in the medium and consequently, a spatial variation of the refractive index, that acts as a thermal lens resulting in phase distortion of the propagating beam.

The diffraction efficiency studies on the polymer films are studied through grating formation. All the polymers were investigated for grating formation using diode pumped Nd:YAG laser of wavelength 532nm and He-Ne laser was used as a read out beam.

The application of the nonlinear properties of these dyes as optical limiters are studied. Optical limiting has been studied based on sample position, concentration, medium and aperture size.

The summary of the results, conclusion and suggestions for further work are further discussed.