ABSTRACT

Nonlinear optics has become a very important subfield of optics since its inception over three decades ago. In linear systems, the induced polarization is proportional to the electric field, and the dielectric constant, the optical susceptibility are constant for each particular medium, independent of the electric field. However, when the intensity of the light propagation through the material is increased, the above conditions cease to be constant and nonlinear effect occurs. The synthetic work of scientist during the same period has resulted in abroad range of new materials whose optical and nonlinear optical properties have been characterized. Many of these new materials are organic. Organic materials have emerged during 1980s as an important class of nonlinear optical material that offers unique opportunities for fundamental research as well as for technological applications.

In this thesis work, the nonlinear optical properties of nominated laser dyes in liquid and solid form have been investigated by using four different nonlinear studies. In the first study, six dyes namely acid red 94, acid red 92 and solvent red 45 from Fluorone subgroup and acid red 1, acid red 29 and acid orange 10 from Azo group are selected and explored for their nonlinear optical properties by using single beam method (Z-Scan technique).

In the second study, acid red 94 and acid red 1 doped gelatin films are prepared and optical phase conjugation using degenerate four wave mixing concept are investigated in these films. In the third, in dye doped gelatin thin films holographic gratings are formed and their diffraction
efficiencies studied. Finally, investigations of the optical power limiting capabilities in eight dyes from three different groups at different solvents and concentrations are presented.

In order to explore the nonlinear optical characterization of fluorone subgroup and azo dyes z-scan technique employed. Six dyes have been chosen, three from each group. Since the UV-Vis spectra of five dyes show strong absorption band in the region of 530 - 550 nm, SHG of cw Nd-YAG laser (diode pumped) at wavelength of 532 nm is used in this study. Ar ion laser at wave length of 488 nm is used with acid orange 10 since this dye has absorption peak at 480 nm. In addition to the dye samples in solution form, dye doped polymer rod of thickness ~1mm of concentration 0.2 mM are fabricated for two fluorone dyes (acid red 94 and solvent red 45) in PMMA with high optical quality.

Self-defocusing i.e. negative nonlinearity is observed in all dyes (in both liquid and solid forms). The-defocusing effect is attributed to the thermal nonlinearity resulting from absorption of radiation at 532 nm. Localized absorption of a tightly focused beam propagating through an absorbing dye medium produces a spatial distribution of temperature in the dye sample and consequently, a spatial variation of the refractive index, that acts as a thermal lens in severe phase distortion of the propagating beam.

It is also observed that the nonlinear absorption coefficient for all the dyes can be attributed to a saturation absorption process. The nonlinear refractive index and nonlinear absorption coefficient are found to depend on the surrounding medium parameter, especially on solvent polarity (in liquid form) and the concentration of the dye. The nonlinear refractive index,
nonlinear absorption coefficient and magnitude of the third-order nonlinear susceptibility $\chi^{(3)}$ are calculated for all these dyes.

Organic dyes namely, acid red 94 and acid red 1 from fluorone and azo groups respectively in the form of dye doped gelatin thin films are chosen for the optical phase conjugation study via degenerate four wave mixing. The gelatin films have been sensitized by doping them in different concentrations of the dye solutions for constant time. The phase conjugation signals are produced in the dye doped gelatin films by using SHG of cw Nd-YAG laser at wavelength of 532 nm. The phase conjugate reflectivity (R %) is found to depend on various parameters, such as the concentration of the dye in the gelatin film, the intensity of the probe beam, the intensities of the forward and backward pump beams and the angle between the probe and forward pump beams. Maximum phase conjugation reflectivity of 0.153% and 0.137% has been observed in acid red 1 and acid red 94 respectively. Saturable absorption is the underlying mechanism involved in the observation of optical phase conjugation in the investigated media.

The formation of grating as well as the diffraction efficiency measurements studies are carried out on the dye (acid red 1, acid red 94, acid red 92 and acid red 29) doped gelatin films. Since the four organic dyes show strong absorption around 530-550 nm, the output of SHG of cw Nd-YAG is used for the formation of gratings. When the dye doped gelatin film is exposed to an interference pattern due to two beams of Nd-YAG laser, a permanent surface relief grating formation is observed. The grating formation is monitored using He-Ne laser at 632.8 nm as probe beam and the intensity of the first order diffraction beam is measured as a function of time using detector attached with power meter.
The diffraction efficiency is found to depend on various parameters, such as the concentration of the dye in the gelatin film, the intensity ratio of the writing beams and the spatial frequency. It has been found that the acid red 1 (Azophloxine) sample is more efficient among the four samples with a diffraction efficiency of about 1.319%. A model has been proposed to explain the mechanism of formation of surface relief grating. The exposed region of the films is examined with atomic force microscope (AFM). AFM recording of the exposed interference pattern reveals the creation of surface relief grating. This shows that recording is in the form of thickness modulation as well as amplitude modulation.

The applications of the nonlinear properties of eight dyes from three different groups as optical power limiters are studied under low power cw laser excitation. The mechanism responsible for optical limiting is mainly attributed to the thermally induced nonlinear refraction. The defocusing effect observed in these samples under cw illumination is utilized to demonstrate their optical limiting action. Based on their nonlinear refractive index, the samples behave as good optical limiters even at low powers. These results are quite encouraging for possible applications in nonlinear optical devices. Limiters based on nonlinear absorption phenomena have been designed for use with high power pulsed laser sources, but limiters based on thermo optic nonlinearity such as the one studied here can be used as efficient limiters in the low power cw regime.