CHAPTER 6

SUMMARY AND CONCLUSION

6.1 SUMMARY

There has been a large need for nonlinear optical materials that can be used with low intensity lasers for applications such as phase conjugation, image processing, and optical switching. Large nonlinear optical susceptibility resulting from the nonlinear response of organic molecules has attracted much attention. Many reports have been published for the case of single crystals of organic molecules, organic molecules in liquid solutions, and organic and biologic materials doped in various solids.

In the first study, the third-order nonlinear optical properties of triphenylmethane dyes (basic green 1 and acid green 5) and oxazine dyes (nile blue chloride and nile blue A) have been measured by the Z-scan technique. Z-scan method has become a standard tool for measurement of the nonlinear absorption and nonlinear refractive index coefficients of various organic materials. Z-scan is a simple and sensitive method, in which both the sign and type of nonlinearity of the material can be explored immediately after the measurement is finished.

In the second study, organic dyes embedded in solid matrices are used as nonlinear medium to generate optical phase conjugation signal. Triphenylmethane dyes (basic green 1 and acid green 5) sensitized gelatin films are used to carry out the proposed work.
Acid orange 6 and acid orange 10 from azo family are chosen in the third study. Dye doped thin films are formed and self-diffraction studies are carried out in these films. Self-diffraction, one of the nonlinear phenomena, is the qualitative study of material properties. In self-diffraction technique, nonlinearity of any organic material is explored by testing the grating formation capability of the material either in liquid form or in solid form and studying diffracted beams of different orders.

Holographic data storage technology is an emerging new field of information storage. Data storage has gained importance as new materials started developing. Recently, polymeric materials have been widely used for holographic data storage. In the fourth study, sudan dye based poly (methacrylate) films are used as a holographic recording media. A brief summary of the results of the investigation is presented.

In the present investigation, all the experimental studies are repeated four times to verify the reproducibility of the results and after confirming the consistency, the results are presented. The estimated errors do not exceed ±5%.

6.1.1 Nonlinear Optical Properties of Triphenylmethane and Oxazine Dyes

In this, four dyes namely basic green 1 and acid green 5 from triphenylmethane family and nile blue chloride and nile blue A from oxazine family are chosen. Since the dyes chosen show a strong absorption around 625 nm and 639 nm, 632.8 nm wavelength of He-Ne laser is used for the study. The nonlinear optical properties of aqueous solution of triphenylmethane dyes and solution of oxazine dyes are investigated using Z-scan technique. The Z-scan technique is performed by sending an axially
symmetric beam through a converging lens, and then through the nonlinear optical sample which is placed near the beam waist. The transmittance of the nonlinear medium is measured through a finite aperture placed in the far field as a function of the sample position \((Z)\), measured with respect to the focal plane of the converging lens. The measurements of nonlinear absorption coefficient \(\beta\) and nonlinear refractive index \(n_2\) for aqueous solutions of basic green 1 and acid green 5, and solutions of nile blue chloride and nile blue A in solvent ethanol are carried out using open and closed aperture Z-scan techniques, respectively.

Investigations are carried out to study the dependence of nonlinear absorption coefficient \(\beta\) and nonlinear refractive index \(n_2\) on the concentration of dye and incident beam intensity. It is observed that the nonlinear absorption coefficient and nonlinear refractive index increases with increase in the concentration of dye and incident intensity for the solutions of the triphenylmethane and oxazine dyes. It is observed that the nonlinear absorption coefficient for aqueous solutions of basic green 1 and acid green 5 can be attributed to saturation absorption process while in nile blue chloride and nile blue A in solvent ethanol, it is due to reverse saturation absorption process. Self-defocusing i.e. negative nonlinearity is observed in these dyes. The defocusing effect is attributed to the thermal nonlinearity resulting from absorption of radiation at 632.8 nm. Localized absorption of a tightly focused beam propagating through an absorbing dye medium produces a spatial distribution of temperature in the dye solution and consequently, a spatial variation of the refractive index, that acts as a thermal lens resulting in severe phase distortion of the propagating beam.

The nonlinear refractive index and nonlinear absorption coefficient are calculated for all these dyes. The Z-scan measurements indicate that these
dyes exhibit large nonlinear optical properties. All these experimental results show that these dyes are promising materials for applications in nonlinear optical devices.

6.1.2 Optical Phase Conjugation in Triphenylmethane Dye Films

Optical phase conjugation by DFWM has been demonstrated in triphenylmethane dye (basic green 1 and acid green 5) doped gelatin films. Since these dye doped gelatin films have absorption peak around 625 nm and 639 nm respectively, the beam from He-Ne laser operating at 632.8 nm is used in the DFWM geometry. The output beam is first split by a beam-splitter BS1 (5:90). The beam reflected off from BS1 is used as probe beam $E_3$ after reflection by beam splitter BS3. The transmitted beam from BS1 is further divided by another beam-splitter BS2 (50:50), called forward pump wave $E_1$ and backward pump $E_2$, respectively. Beam-splitter BS3 (50:50) is used to direct the probe beam to the dye doped gelatin film, and to transmit the PC signal, which is opposite to the direction of the probe beam.

The phase conjugate reflectivity is found to depend on (1) time of exposure (2) the concentration of the dye in the gelatin film (3) the intensity of the probe beam (4) the intensity of backward pump beam (5) the intensity of forward pump beam and (6) the interbeam angle between the probe and forward pump beams.

It is found that the phase conjugate reflectivity increases linearly with time till it reaches maximum value and after that it decays very slowly. Phase conjugate reflectivity is studied for different concentrations of the dye in the gelatin film. It is observed that the PC reflectivity increases with concentration of the dye doped up to certain concentration and then decreases.
for higher concentrations (lower transmittance). It is also observed that the PC reflectivity increase with increase in intensities of forward, backward and probe beams. The PC reflectivity is studied for 6° to 18° range of interbeam angle between the probe and forward pump beams. It is found that the PC reflectivity first increases and then decreases.

Comparison of result indicates that acid green 5 doped gelatin films have higher reflectivity than the basic green 1 dye doped gelatin films. The maximum reflectivity achieved is 0.12% for acid green 5 plates with 43% transmittance and 0.10% for basic green 1 plates with 44% transmittance. The time for the maximum phase conjugated reflectivity is of the same order for both dyes, being approximately 3 minutes. These reflectivities are larger than those obtained with other dye doped gelatin films.

6.1.3 Self-diffraction Study in Azo Dye

The azo dyes (acid orange 6 and acid orange 10) soaked gelatin films used in this study show maximum absorption at 495 and 480 nm. Hence 514.5 nm beam from argon ion laser is used for grating formation. Exposure of these dyes soaked gelatin films to interfering beams of argon ion laser results in the formation of transient self-diffraction gratings. The diffraction efficiency of the grating depends on the same parameters as mentioned for azo dyes. Rate of grating formation is monitored by measuring the intensity of first order diffracted beam.

It is found that the diffracted intensity in the first order increases linearly with time till it reaches saturation. The intensity begins to decrease after sometime and decays completely within 1.5 hours i.e. the grating is fully erased by further exposure of the interference pattern. The formation of
grating is studied for different powers of the writing beams. It is observed that higher the power of the writing beam, greater the rate of grating formation.

Experiments are performed with three different intensity ratios (50/50, 60/40, 70/30) of the interfering beams. Studies have shown that the rate of formation of grating mainly depends on intensity ratio. It is found that for all the films with different concentrations, the grating formation is immediate and the diffracted intensity rises linearly to a maximum until saturation is reached. Also it is found that in the films of lower concentration, the grating formation is not very efficient when compared to that of the grating formed in the films of higher concentration. These results confirm that the grating formed increases with the increase in the concentration of the dye. The dependence of the first order diffracted power on the angles between the interfering beams is also investigated. We observed that the rate of formation of grating decreases with increase in the angle between the interfering beams.

Beam blocking study is carried out while the formation of grating is in progress. Blocking one of the beams while writing is in progress before saturation is reached has resulted in decrease in diffracted intensity whereas after saturation is reached the blocking has resulted in increase in diffracted intensity. It is found that the different orders of diffracted beams are not in phase with each other and also the phase difference varies with grating formation. Three diffracted order is observed in acid orange 6 doped gelatin films.

6.1.4 Characterization of Sudan Dye Based Poly(Methacrylate) Films

Since the Ia, Ib and Ic polymers have absorption peak around $517 \pm 1$ nm and IIa, IIb and IIc polymers have absorption peak around $510 \pm 1$ nm, argon ion laser is used for the formation of grating. Exposure to
interference pattern created due to superposition of two beams from the laser results in permanent grating and the formation of the grating is monitored by measuring the intensity of the first order diffracted beam using a He-Ne laser beam at 632.8 nm. The formation of grating strongly depends on the time of exposure, irradiation intensities of writing beams, intensity ratios of the writing beams, the concentration of polymer in the film and angle between the interfering beams. From the investigations carried out the following conclusions are drawn.

The diffraction efficiency increases linearly with time of exposure until saturation is reached. The rate of formation of the grating is studied for 40 to 70 mW range of power of each writing beam. For Ia, Ib, IIA, IIB and IIC polymers films it is found that with increase of the power of the writing beams the formation rate also increases while it is found to decrease for Ic polymer. This may be attributed to the less thermal stability of Ic polymer. Also grating formation rate depends on the intensity ratios of the writing beams, being maximum for 50/50 intensity ratio. It is also observed that the diffraction efficiency of the grating increases with increase in the concentration of the polymers. Also the study is carried out for varying angles between the interfering beams. It is found that the formation of grating decreases with increase in the angle between the interfering beams. Comparison of results indicates that Ib polymer films have higher diffraction efficiency than all the polymers for same recording parameters. Maximum diffraction efficiency achieved is 1% for Ib polymer films and three diffracted orders are observed.

Examination of the recorded region by an optical microscope clearly shows the formation of the grating structure. Diffraction efficiency in the film has remained constant over a long time period in this material.
Atomic Force Microscopic investigations of a grating structure recorded in Ib polymer films reveals the creation of regularly spaced sinusoidal surface relief structures with a period of 2.6 μm which is consistent with the optical microscopic observation. The depth of the surface relief patterns is about 41.15 nm for the 9 wt% concentration of Ib polymer. The properties observed so far make the gratings (materials) promising one for use as optical storage media using Fourier holographic technique.

6.1.5 High-Density Optical Data Storage

High-density optical data storage using Fourier holographic technique is carried out in Ib polymer films. One full page of text to be stored is converted into transparent slide using photographic technique and the transmitted light called the object beam is made to interfere with the reference beam at a well defined position in the Ib medium. The resulting interference pattern at the focused spot of size 50 μm produces the permanent grating. When the recording of one page is completed, a different position on the storage medium is addressed for recording of second page. Finally, the entire storage medium is covered with an array of holograms each containing a data page. Since the information is stored in the form of Fourier transform hologram in the focused spot without binary conversion, the data storage capacity is very large. In a 5 cm dia circular disc $10^5$ pages in the form of text, figures, drawings etc., can be stored without multiplexing.

For read-out of the stored page, the desired hologram is addressed by the reference beam of He-Ne laser. The light diffracted from the hologram is a faithful replica of the signal wavefront from the page recorded.
6.1.6 **Salient Features of the Present Investigations**

The following are the salient features of the present investigations:

- Using the Z-scan technique, the nonlinear refractive index and absorption coefficients and third-order susceptibility are explored in aqueous solution of triphenylmethane dyes and they are found to have high nonlinear coefficients.
- In oxazine dyes, the nonlinear refractive index and absorption coefficients and third-order susceptibility are explored in solution of these dyes in solvent ethanol and they are found to have high nonlinear coefficients.
- Low power optical phase conjugation is demonstrated in triphenylmethane dyes doped gelatin films.
- In azo dye doped gelatin films self-diffraction grating is formed.
- Using Fourier transform holographic method high-density optical data storage is demonstrated in Iβ polymer films.

6.2 **SUGGESTIONS FOR THE FUTURE WORK**

- To select organize dyes with structures favorable for nonlinear optical property and synthesis of dye hooked polymers for nonlinear studies.
- Apply these nonlinear polymers in device fabrication.