A Review on Organic Materials for Optical Phase Conjugation & All-optical Switches

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ABSTRACT

Nonlinear optical materials with optimum properties are essential in continuous development of photonic and electro-optical devices, used in optical communications, networking, optical computation for signal processing, and data storage equipments. The changing trend is to use organic materials/dyes that exhibit exceptional nonlinear optical properties instead of conventional materials which have comparatively low nonlinear properties. These organic materials/dyes are easy to prepare in solution or solid form. The resulting organic material has a low dielectric constant, eliminating the need for poling while maintaining the refractive index. However, these organic materials have few of the drawbacks inherent in the processing of comparable inorganic materials like of intense light induced degradation or bleaching and aggregation at higher dye concentration. In order to overcome these drawbacks and for effective use of highly nonlinear dyes, the dye molecules are doped in polymer matrix. This idea of dye-doped polymer material matrix may increase the concentration of absorptive or fluorescence centers as well as the opto-chemical and opto-physical stability. In this paper, we have discussed the strategic advantages of dye-doped polymer nonlinear materials in comparison with organic and inorganic nonlinear materials for optical phase conjugation and all optical switches in future photonics technology. This was done by studying the linear optical properties and nonlinear optical Phase Conjugation properties of two azo dye-doped polymer films by considering organic dyes disperse orange (DO-25) and disperse yellow (DY-7) doped in a polymer matrix Polymethyl methacrylate methacrylic acid (PMMA-MA). The nonlinear optical phase conjugation properties are studied using Degenerate Four Wave Mixing set-up using 532 nm wavelength CW laser beam. The effect of dye concentration, intensity of backward, forward pump, and inter beam angle between

probe and forward pump beam on phase conjugation reflectivity are also studied and compared.

Keywords : Azo Dye-doped Polymer films, Degenerate Four Wave Mixing, Nonlinear Optical Properties, Optical Phase Conjugation.

I. Introduction

Materials with exceptional nonlinear optical properties are critical to the continuing development of photonic and electro-optical devices, such as those used in optical communications, networking, Optical computation for signal processing, and data storage equipments. The nonlinear optical material is a general term for the materials efficiently makes appearance of nonlinear phenomenon optically as the responses to optical wavelength conversion, optical amplification as well as the refractive index changes due to its intensity. Nonlinear optical materials are largely divided into inorganic and organic materials. In 1930, the nonlinear optical effect related to optical wavelength conversion was predicted, which was said to be the first finding knowledge about the nonlinear optical phenomenon. In 1960, laser oscillation using inorganic material was reported. Since then researches of inorganic nonlinear optical materials were actively taken place, but now-a-days, probably there is no more that undiscovered (Shen Y. R., 1975).

With the rapid development of modern science and technology, information transmission capacity of communication increases day by day. Optical communication, which has advantages of large transmission capacity, high transmission velocity, excellent anti-jamming ability and good Signal-to-Noise value, is becoming a main method in communication researches at present. Functions like optical switching and memory by non-linear optical effects, all depending on light intensity, are expected to result in realization of a pivotal optical device in optical computing. This is a new data processing system that makes the maximum use of light characteristics such as parallel and spatial processing capabilities and high speed.

All-optical networks with good performances, such as big capacity, good transparency, wavelength routing characteristics, compatibility and extensibility, has become the first choice of next generation of wide-band net with a promising application. Accompanied by the deep research of wave division multiplex (WDM), switches have drawn more and more people's attention. In the existing optical-electronic-optical conversion apparatus of present communication net, disadvantages of slow switching speed and clock displacement have lead to a "bottleneck" of optical fiber communication systems. All-optical switches which can break through the transmission speed limits of electro-optical, acousto-optical, thermo-optical and micro-electro-mechanical switches, can serve as effective methods to solve these problems. Based on the third-order nonlinear optical (NLO) effect, phase all-optical switches use a controlled light to bring changes of refraction index and make phase difference when signal light passes through sample and thus carry out the function of "on" or "off' of optical switches. Its nonlinear phase difference is proportional to $(2\pi/\lambda)n_2IL$, where I is intensity, L is length of interaction of wave and n_2 is nonlinear refraction index. The properties, such as change speed, intensity loss, sensitivity to optical polarization and insert loss, all depend on third-order NLO materials used to synthesize apparatuses. At present, it is with great enthusiasm to emphasis on exploring and synthesis of materials for all-optical switches based on the continual discovery of varies kinds of new materials. There are other applications of third-order NLO materials, including optical limiting devices, Q-switch, passive mode locking, optical operation and light storage etc.. Laser weapons applied to military have special effects on optical-electro antagonism, aerial defense and military recovery. Laser blinding can make eyes blind temporarily or permanently, and laser can also destroy important apparatus in the satellite, such as detectors and sensors. As a result, laser protection materials and devices have become a focus. The purpose of laser protection is to protect people and devices from damage of high intensity. These optical limiting devices are mainly based on the materials' third-order NLO properties, including self-focusing, self-defocusing, two-photon absorption, reverse saturable absorption and nonlinear scattering. Comparing to earlier laser protection devices, it has advantages of fast response, wide protected band, low optical limiting threshold, large damage threshold and high linear transmission, etc. The third-order NLO properties of materials can also be used in the compression (mode-locking) and shaping of laser pulses, optical bistability, etc.. Third-order NLO materials also have many potential practical exciting applications, and motivated scientists to continually explore new materials with high third-order NLO properties. The demands of materials for all-optical information process and high-speed all-optical switches include large nonlinear refraction index, small linear and nonlinear absorption coefficient, fast response and low propagation loss (Van Stryland and Chase, 1994, Hutchings and Van Stryland 1992).

Photonic and electro-optical (in which information storage or processing involves the modulation and switching of light beams) devices are used in many applications which include : Electro-optic modulators , Mach-Zehnder interferometers, Optical switches, Optical interconnectors, Frequency doublers for high-power lasers, Active waveguides, Optical memory storage devices, Optical computing devices, Nonlinear directional couplers, Nonlinear Bragg reflectors, Optical limiters, Photorefractive memories. The global market for Non-Linear Optical Materials and Applications is in Millions of US$. There are several companies including many key and niche players worldwide such as CASIX, Inc., Cleveland Crystals, Inc., Coherent, Inc., Conoptics, Inc., Cristal Laser SA, Crystal Technology, Inc., Deltronic Crystal Industries, Inc., EKSMA OPTICS, Fujian Castech Crystals, Inc., Inrad Inc., JDS Uniphase Corporation, Laser Optics, LINOS Photonics GmbH & Co. KG, Northrop Grumman Synoptics, Nova Phase, Inc., Quantum Technology, Inc., Raicol Crystals Ltd., Saint-Gobain Crystals, and Vloc, Inc. template, modified in MS Word 2003 and saved as "Word 97-2003 & 6.0/95 – RTF" for the PC, provides authors with most of the formatting specifications needed for preparing electronic versions of their papers. All standard paper components have been specified for three reasons: (1) ease of use when formatting individual papers, (2) automatic compliance to electronic requirements that facilitate the concurrent or later production of electronic products, and (3) conformity of style throughout a conference proceedings. Margins, column widths, line spacing, and type styles are built-in; examples of the type styles are provided throughout this document and are identified in italic type, within parentheses, following the example. Some components, such as multi-leveled equations, graphics, and tables are not prescribed, although the various table text styles are provided. The formatter will need to create these components, incorporating the applicable criteria that follow.

II. Nonlinear Materials : New Initiatives

Currently, there exists a wide range of inorganic non-linear optical materials (Giuliano and Hess, 1967, He et. al. 1997) with varied wavelengths, damage thresholds and optical characteristics. The research focus is to develop materials that meet all requirements such as faster response, high laser damage threshold and wide transparency range coupled with adaptability, processing ability and the ability to interface with other materials. Further robust growth in demand for high bandwidth fiber optic networking infrastructure and high speed optical computing are expected to boost the demand for Non-Linear Optical Materials.

On the other hand, the 1982 ACS symposium report added momentum to the organic nonlinear optical material studies. Since then the studies have started to become active. It revealed organic compounds with the delocalized conjugated electrons which have excellent nonlinear optical property and high-speed responsiveness due to high mobility of electrons. The 21st Century is said to be an age of photonics. As one of the basic technology of photonics, improvements of the wavefront control technology using organic nonlinear optical effects are considered very important. Now R&D on the organic materials with excellent nonlinear optical properties, and vigorous applied studies have been carried out.

Present developments in the field of materials chemistry show that, though inorganic materials are still the choices for many devices, interest in organic materials are growing day-by-day in view of their adaptability to various kinds of applications. The field of organic molecular materials has transformed the use of materials in the modern world in the last 20 years, and it can be seen that organic molecules provide wonderful opportunities to materials researchers to design custom-tailored materials whose properties at the macroscopic /microscopic level reflect closely to the modeled or actual behavior of individual molecules. In other words, development of novel functional organic materials is a rapidly growing area of science, which probably can replace the traditionally used materials with cheaper and better-performing new ones in the near future, and also bring out some new applications (Olga et. al. 1998, Mukherjee et. al. 1997, Perry 1997, Wei 1992, Swalen and Kajzar 2001).

In view of the technological applications of the organic materials, the current research focus is in five technical areas, which are (1) Structural and multifunctional materials, (2) Energy and power materials, (3) Photonic and Electronic Materials, (4) Functional organic and hybrid materials, (5) Bio-derived and bio-inspired materials. Among the five main technological thrust areas of the organic materials, the focus of this thesis is on “PHOTONIC MATERIALS”, which can find applications in the field of linear and electric nonlinear optics (otherwise known as photonics). Some of the benefits of organic nonlinear optical materials are :

- **Easy to process**: Because they do not require electric poling or the preparation of large single crystals, these materials are easier to process than inorganic optical materials.
- **Lower cost**: The ease of processing directly translates into a lower cost to fabricate.
- **High second- and third-order susceptibility**: This technology exhibits exceptional performance in doubling and tripling the frequency of light passing through it, making it at least comparable to inorganic materials.
- **Low dielectric constant**: An optical material with a high dielectric constant requires a larger poling voltage in order to polarize the dipole moment and can suffer changes in the

refractive index. This technology requires no poling voltage and maintains its refractive index.

- **High electro-optic coefficient**: Materials with a high electro-optic coefficient are more suitable for electro-optic modulation for high-speed devices.
- **Colourless**: It is believed that the clarity of the doubling material will prevent the absorption of visible light, allowing a wide variety of light frequencies to be doubled.
- **Resistant to laser damage**: The tripling material can be exposed to 432,000 20-nanosecond pulses at 20 Hz without any evidence of damage to the organic material, making it ideal for use in photonic applications.

These organic materials have few of the drawbacks inherent in the processing of comparable inorganic materials like of intense light induced degradation or bleaching and aggregation at higher dye concentration. In order to overcome these drawbacks and for effective use of highly nonlinear dyes, the strategic idea for the next practice is doping the dye molecules in polymer matrix. This idea of dye-doped polymer material matrix may increase the concentration of absorptive or fluorescence centers as well as the opto-chemical and opto-physical stability (Fisher 1983, Yariv 1978).

Nonlinear optical phase conjugation (OPC) by degenerate four-wave mixing (DFWM) is an important technique with applications in many fields of science and technology including image transmission, optical image processing, optical filtering, and laser resonators (Fisher 1983). When two counter propagating and intense light beams interact with a nonlinear medium, together with a less intense third one, a fourth beam is generated from the medium, which will be the phase conjugation of the third beam. This technique is called four-wave mixing. The unique feature of a pair of phase-conjugate beams is that the aberration influence imposed on the forward (signal) beam passed through an inhomogeneous or disturbing medium can be automatically removed from the backward (phase-conjugated) beam passed through the same disturbing medium (Yariv 1978). The main applications of degenerate DFWM techniques are nonlinear spectroscopy, real time holography, and phase conjugation. Phase conjugation by (DFWM) has been demonstrated in many organic or inorganic materials using pulsed or continuous-wave (cw) lasers (Tanaka 2002, Geethakrishnan and Palanisamy, 2006).

The organic molecules exhibit large polarizabilities because exited π-bond electrons are delocalized and hence easily polarizable. Nonlinear absorption like two photon absorption and saturable absorption plays a very important role when dyes are used for the production of phase conjugation light, because χ^3 is inversely proportional to the saturation intensity. These systems exhibit large third-order susceptibilities. OPC has been reported in Glasses and other solid matrices doped organic dyes emerged as promising materials for OPC because of their large third-order nonlinear susceptibilities χ^3. In these materials, the phase-conjugate wave can be generated at low light intensities provided by the continuous-wave lasers. Moreover, these materials can be easily prepared in the laboratories. The important fundamental physical processes like nonlinear refraction, thermal grating, saturation and reverse saturable absorption, two photon induced fluorescence, photorefraction, and stimulated brillouin scattering etc. may lead to the formation of a laser-induced grating in the medium are associated with the generation of phase conjugated wave (Fisher, 1983).

In this paper we have reviewed the linear optical properties and nonlinear optical Phase Conjugation properties of two azo dye-doped polymer films by considering organic dyes disperse orange and disperse yellow doped in a polymer matrix Polymethyl methacrylate methacrylic acid (PMMA-MA). The nonlinear optical phase conjugation properties is studied using Four Wave Mixing set-up using 532 nm wavelength CW laser beam. The linear absorption, single photon fluorescence, two photon induced fluorescence behavior are studied. PC Reflectivity as function of angle between the probe beam and forward pump beam and transmittance as a function of time are studied (Shubhrajyotsna et.al. 2013, 2015).

III. Design of Nonlinear Molecule

General Criteria

One of design strategy is proposed recently by Albota et.al. (1998) dealing with molecules based on benzene ring as π-center which is attached symmetrically by either electron-donor (D) or electron-acceptor (A) through various lengths of conjugated connectors; D-π-D or A-π-A. They concluded that σ is increased by increasing the length of conjugation; change with the D/A strength and the extent of symmetric intramolecular charge-transfer (CT) from the D ends to the π-center or vice versa, meaning that symmetric charge redistribution effectively occurs upon excitation of such symmetric molecules. A similar approach was made in designing molecules by Reinhardt (1997) and his coworkers. dealing with benzene ring as π-center which is symmetrically coupled with two electron acceptor (A-π-A) or asymmetrically with D and A (D-π-A), respectively. There is no clear effect of structural symmetry on σ values, although increasing conjugation length of π-center brings about a significant improvement of the value. In fact, an asymmetric structure, D-π-A. This seems to suggest that there must be more crucial molecular factors other than structural symmetry involved. In this study we have considered two azo dye molecules.

Sample Preparation

Commercially available [3-[N-ethyl-4-(4-nitrophenylazo) phynyl-amino]propionitrile (Disperse Orange-25) as shown in Fig. 1 and 4-[4-(Phenylazo)phenylazo]-o-cresol (Disperse Yellow-7) as shown in Fig. 2 (Aldrich Chemical Co.) are purified by recrystallization twice with spectrograde ethanol and by vacuum sublimation. The purity is determined spectroscopically. Purified chloroform is used as the solvent. To prepare the film, Polymethyl methacrylate – metacrylic acid was used as polymer matrix. The thin films of DO-25 and DY-7 doped in PMMA-MA is prepared using hot press technique. Thin films of variable thickness are obtained between two glass slides.

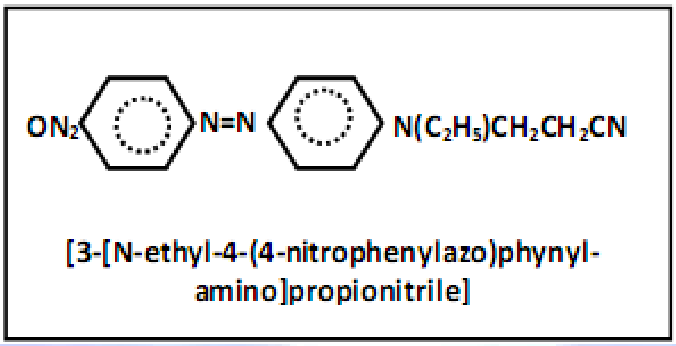

Figure 1 : Molecular structure of Disperse Orange -25.

Linear Optical Properties of DASPB

The linear absorption spectra of DO-25 and DY-7 doped in PMMA-MA are measured on a VARIAN Cary UV-vis-IR recording Spectrophotometer. Fig. 3 and Fig. 4 show the linear absorption spectrum of these samples respectively. The spectral curve has shown that there is a strong absorption band with peak absorption located at 468 nm in case of DY-7 and at 468 nm with a bandwidth of 100 nm, a medium absorption peaked at 270 nm with a bandwidth of 60 nm in case of DO-25 and no linear absorption is observed in entire spectral range of 580 to 1200 nm.

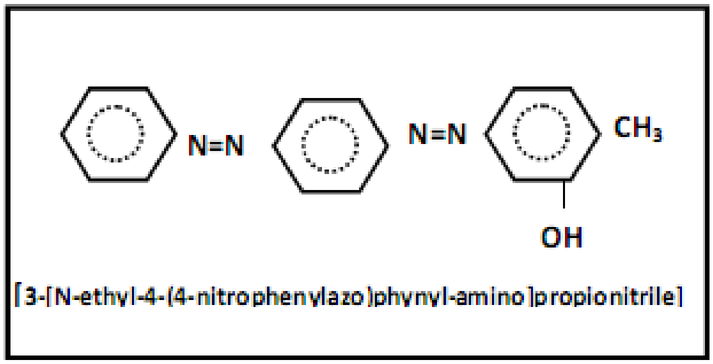

Figure 2 : Molecular structure of Disperse Yellow -7.

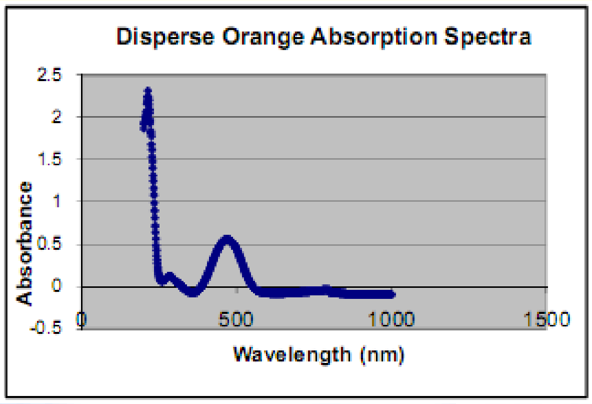

Figure 3 : Linear absorption spectrum of DO-25 in PMMA-MA polymer matrix.

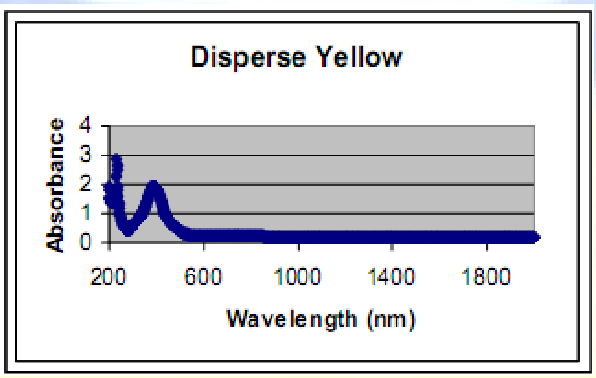

Figure 4 : Linear absorption spectrum of DY-7 in PMMA-MA polymer matrix.

IV. Study of Phase Conjugation

Experimental Configuration for DFWM

The schematic diagram of the phase conjugation experiment is shown in Fig. 5. A CW Nd:YAG laser beam of variable input power at 532 nm was divided into three beams, two counter-propagating pump beams E_1 and E_2 namely forward-pump and backward-pump beams respectively and a probe beam E_3 to form the DFWM configuration. The spot size of each of these three unfocussed beams at the nonlinear medium was 1.0 mm in diameter. The constant power ratio of the probe beam (E_3), forward-pump beam (E_1) and backward-pump beam (E_2) used in this work was ≈ 1 : 10 : 10. The angle between the probe beam and the forward-pump beam was initially 8^o. The sample was exposed simultaneously to all these three beams. The optical path lengths of all the three beams were made equal, so that they were coherent at the sample. The phase-conjugate wave retraces the path in the opposite direction to that of the probe beam E_3 and was detected with the help of a photo detector and power meter. The experimental set-up was mounted on a vibration isolation table to avoid the destruction of the laser-induced gratings formed in the DO-25/DY-7 dye-doped polymer matrix due to mechanical disturbances. The effect of Phase Conjugation signal strength (PC reflectivity) as a function of recording time for different concentrations of dyes doped in PMMA-MA polymer matrix, PC reflectivity as a function of angle between the probe beam and the forward-pump beam, dependence of PC reflectivity on backward pump Intensity by keeping the power of the forward pump and probe beams constant, dependence of PC reflectivity on forward pump power by keeping the power of the backward pump beam and probe beam constant, the conjugate beam reflectivity as a function of the input probe beam intensity, and Transmittance of the sample as a function of time were studied (Shubrajyotsna Aithal et. al. 2011).

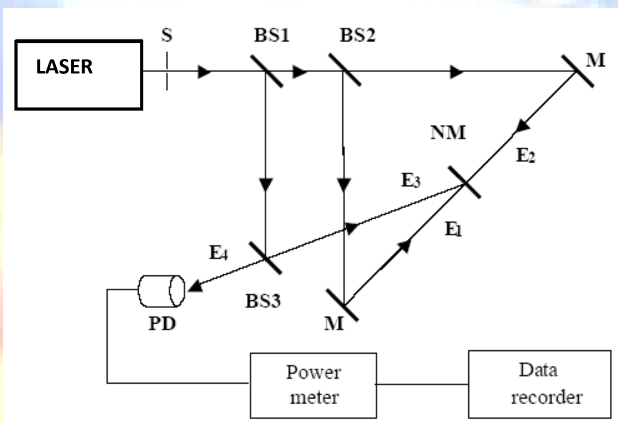

Figure 5 : Experimental set-up for observation of PC wave, S, Shutter; BS1–BS3, Beam splitters; M, Mirror; NM, Nonlinear medium; PD, Photo-detector.

V. Results & Discussions

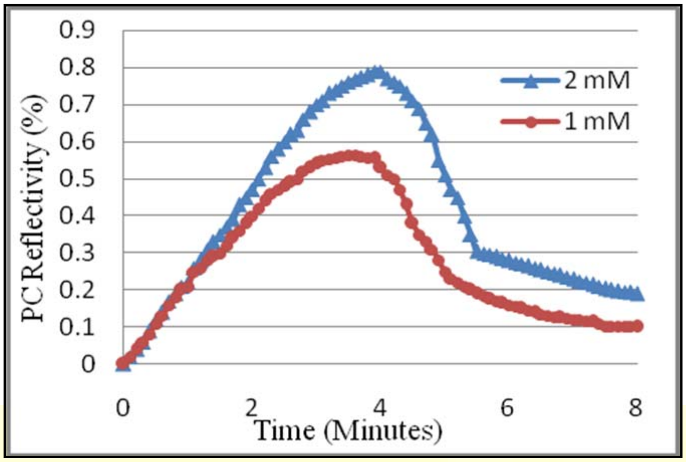

Figure 6 : PC signal versus recording time for different concentration for DO-25.

The PC signal measurements are taken by varying the parameters which influence the PC signal reflectivity during the DFWM process. Fig. 6 shows the PC signal strength versus the time for different dye concentration of the DO-25 doped polymer films and Fig. 7 shows the PC signal strength versus the time for two dye concentration of the DY-7 doped polymer film. It is found that the PC intensity rises linearly to a maximum and then starts decreasing. The phase grating formed is transient. To get maximum reflectivity, it is necessary that there be a perfect overlap of the probe and the pump beams in the nonlinear medium.

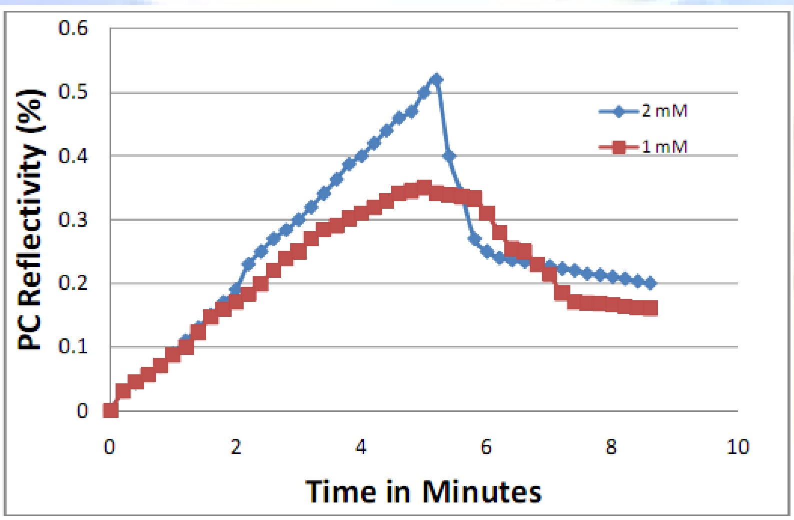

Figure 7 : PC signal versus recording time for different concentration for DY-7.

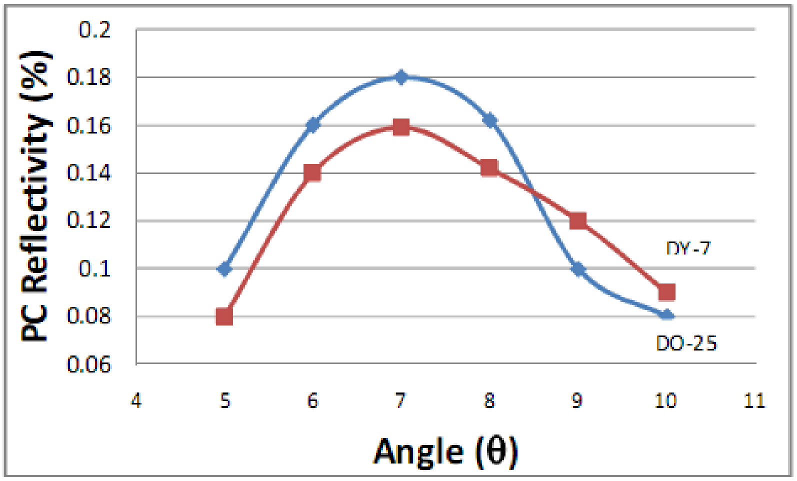

Figure 8 : PC Reflectivity as function of angle between the probe and forward pump beams.

Fig. 8 shows the PC reflectivity as a function of recording angle between the forward pump and probe beam of DO-25 and DY-7 samples. It seems from the figure that, as the angle between the probe beam and the forward pump beam increases, the PC reflectivity first increases and then decreases. This may be because as the angle increases, the probe beam becomes elliptical and only a fraction of its area falls within the interaction region. Because of two-wave coupling, the maximum PC reflectivity is achieved when the angle is 7 degrees in case of both DO-25 and DY-7 samples.

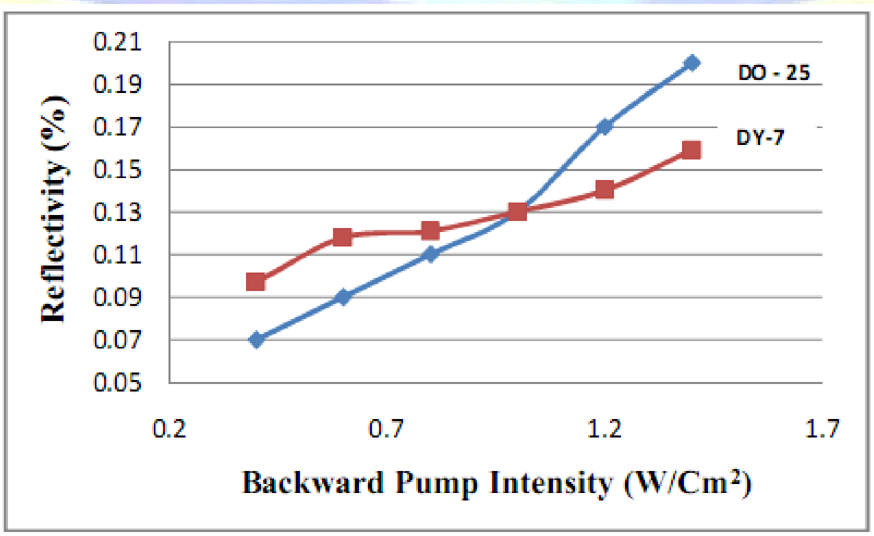

Figure 9 : Dependence of PC reflectivity on backward pump Intensity.

A maximum reflectivity value of 0.18 % is observed in case of DO-25 and of 0.16 % is observed in case of DY-7 for probe beam intensity at 2.5 W/cm^2, and further increase in probe beam intensity resulted to decrease in PC reflectivity. The effect of the backward pump beam power on the PC reflectivity of both the samples by keeping the power of the forward pump and probe beams constant and varying the backward pump beam is shown in Fig. 9.

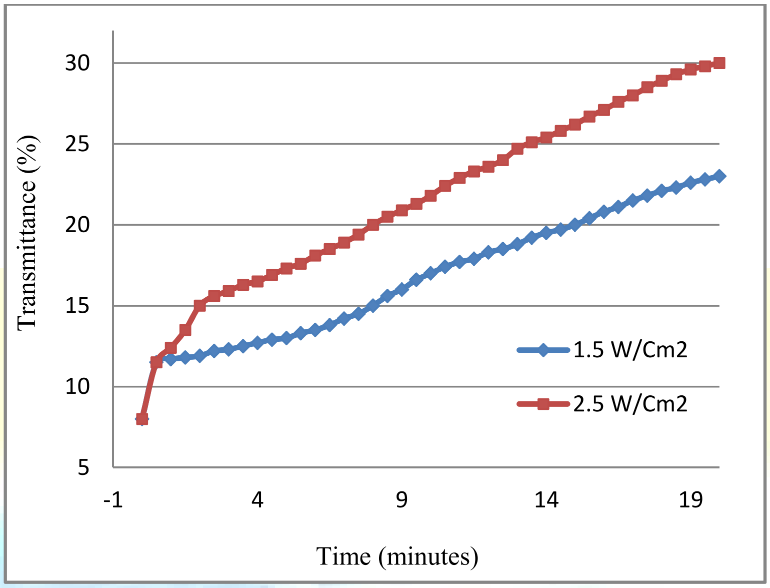

Figure 10 : Transmission as a function of time for DO-25.

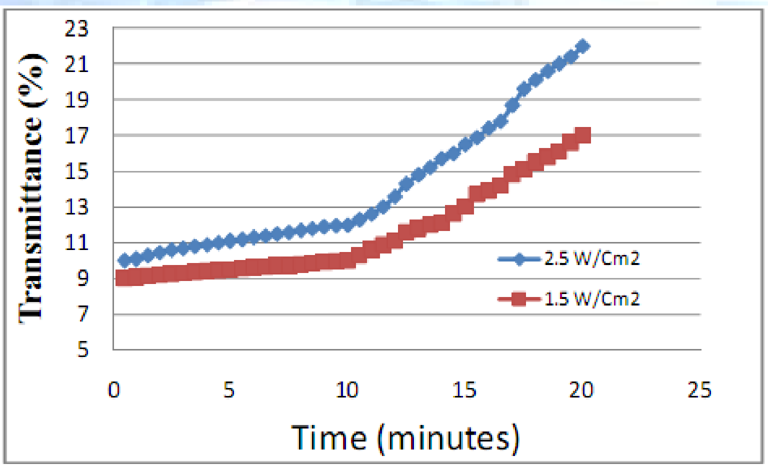

Figure 11 : Transmission as a function of time for DY-7.

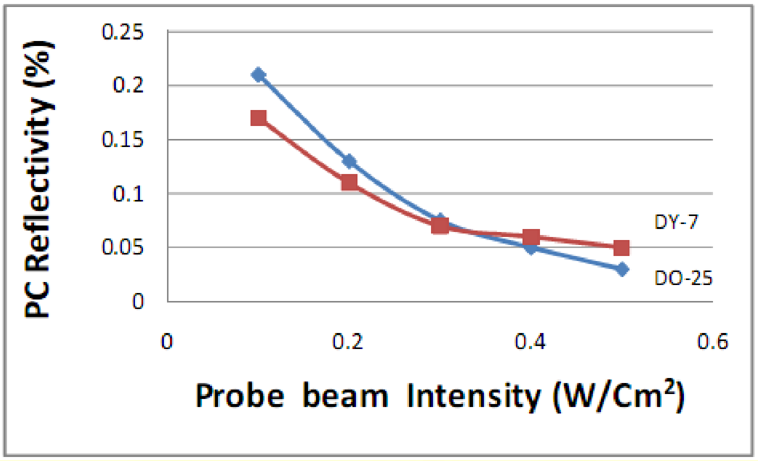

Figure 12 : Conjugate reflectivity as a function of probe beam intensity.

Fig. 12 shows the influence of the input probe beam intensity on the conjugate beam reflectivity. A maximum reflectivity value of 0.22% is observed in case of DO-25 and of 0.17% is observed in case of DY-7 for probe beam intensity at 0.11 W/cm^2 respectively, and further increase in probe beam intensity resulted to decrease in PC reflectivity. Similar observations have been reported in other kinds of material doped with organic dyes (Miniewicz 1997). Fig. 13 shows the variation of reflectivity for different power of forward pump beam for both the samples. The PC reflectivity increases linearly with the power of forward pump beam. There are two main processes which must be considered in the discussion of origin of OPC in dye doped PMMA-PA films: (1) the formation of thermal grating and (2) third order nonlinear optical processes. The DO-25 and DY-7 films illuminated with 532 nm radiation of variable intensity and the transmittance of the sample is measured simultaneously by using photodetector. If the effect observed in our experiments is of purely thermal nature, bleaching of the sample film will be observed. The results obtained for the sample are shown in Fig. 10 and Fig. 11 respectively. It is clearly demonstrated that the transmission of sample increases with time. The experiment described above indicates that the third order nonlinear processes like reverse saturable absorption mainly responsible for OPC in the sample under study.

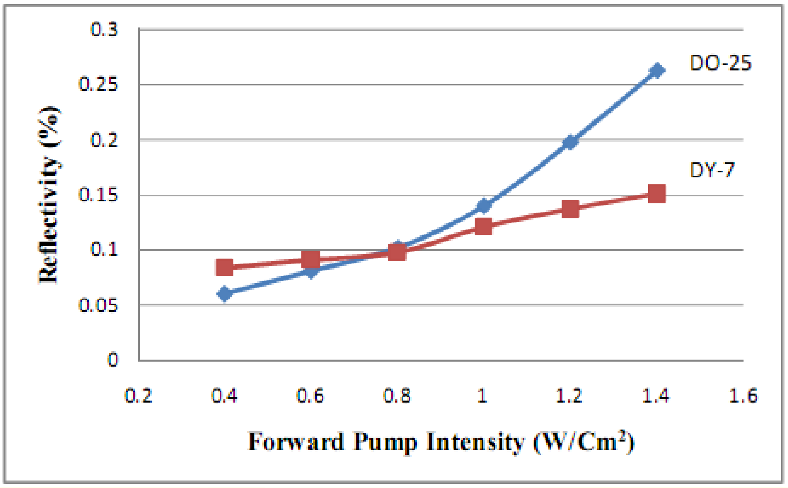

Figure 13 : Dependence of PC reflectivity on forward pump power.

Dyes doped in polymer matrix have the capability of generating a phase-conjugate wave by not only DO-25/DY-7 but also holographic process (Bindhu et. al. 1997). To distinguish the phase-conjugate wave generated by DFWM from that by the holographic process, the transient behavior of the PC signal was studied. For this, the DO-25/DY-7 dyes doped in PMMA – MA polymer matrix were first illuminated with three waves E_1, E_2 and E_3 for a specified duration, and afterwards, E_1 and E_3 were successively turned off, so that only E_2 was incident on the dye film. Fig. 10 and Fig. 11 show the measured phase-conjugate signal as a function of time. The initial rise to a peak within a few minutes is due to DFWM and holographic processes; the sudden drop in the intensity of the PC signal after shutting off both the write beams E_1 and E_3 indicates the contribution from the fast DFWM process. Due to the holographic process the PC signal is present even after E_1 and E_3 are shut off, and it decays rather slowly. If the phase-conjugate wave was generated only by DFWM, the lack of only one of the three beams E_1, E_2 and E_3 would have stopped generation of the phase-conjugate wave. Therefore, it is inferred that the rapidly decaying component corresponds to the phase-conjugate wave which is generated by the DFWM. On the other hand, if spatially modulated information formed by E_1 and E_3 can be recorded in the DO-25 and DY-7 dyes in PMMA – MA polymer film, the phase-conjugate wave can still be generated when E_2 tries to read this stored information, during the lifetime of the holographic grating.

VI. Conclusion

We have observed low-intensity optical phase-conjugation in organic material DO-25 dye in PMMA – MA polymer matrix and DY-7 dye in PMMA – MA polymer matrix using a degenerate four-wave mixing set-up, employing 532 nm light radiation from a CW Nd;YAG laser. The phase-conjugate signal is found to have contributions from the DFWM and the holographic processes. The maximum phase-conjugate beam reflectivity observed in these dye films is about 0.22% in DO-25 doped PMMA-MA matrix and 0.17% in case of DY-7 doped PMMA-MA polymer matrix. The maximum PC reflectivity is achieved when the angle between probe and forward pump beam is 7 degrees. The effects of dye concentration, intensity of backward, forward pump and inter beam angle between probe and forward pump beam on phase conjugation reflectivity are also studied. PC signal strength first increases and then decreases. PC reflectivity is increased by increasing the intensity of the backward and forward pump beam. The polarization and intensity profile are verified to be preserved in the conjugate signal. The predominant phase conjugation signal is attributed to the facts that reverse saturable absorption and large third order susceptibility of the dye molecules. Since the DO-25 and DY-7 dyes in PMMA – MA polymer film are used at 534 nm and this may be suitable for low-power semiconductor lasers in the red wavelength region, DO-25/DY-7 dyes in PMMA – MA polymer film may be a promising organic material for real-time double-exposure phase-conjugate interferometry.

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