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**Predoctoral Training Grant in the Area of
Physical Sciences**

Final Technical Report

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Putcha Venkateswarlu
Department of Physics
Alabama A&M University
Huntsville, Alabama 35762

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PROJECT IN CHARGE/DIRECTOR:

1. DR. M. C. GEORGE

FROM : 9/30/1989

TO: 8/31/1990

2. DR. P. VENKATESWARLU

FROM : 9/1/1990

TO: present

PREDOCTORAL TRAINEES:

1- HOSSIN A. ABDELDAYEM	1989-1990
2- MOSTAFA DOKHANIAN	1990-1991
3- Yao G. Ding	1991-1992
4- BO SU CHEN	1991- present
5- MOHAMMED MAHDI	1992 - present

INTRODUCTION

This final technical report represents the results of the research in nonlinear optics (optical phase conjugation) obtained by five (5) predoctoral students in the department of physics at Alabama Agricultural and Mechanical University (AAMU). Two of these five students have already been graduated. Two are currently working under this grant.

The first section of the report summarizes the results obtained by Dr. Hossin Abdeldayem who graduated in December of 1990. Dr. Abdeldayem has investigated on the nonlinear optical characteristics of organic materials both in solutions and glass form. He studied Disodium fluorescein doped in boric acid glass and 7'- dicyno-apocarotene in hexane and Chinese tea. To estimate the third order nonlinear susceptibilities of these organic materials, he used three different nonlinear techniques. These are Degenerate Four Wave Mixing, Self-Trapping, and Spatial Self-Phase Modulation. The studies had indicated that disodium flurescein doped boric glass was a good optical phase conjugator.

The second section of the report is concerned with the results of the reserch in nonlinear optics performed by Dr. Mostata-Dokhanian who also graduated in May of 1991. Dr. Dokhanian's Predoctoral research was concentrated on the applications of nonlinear photorefractive BaTiO_3 crystal in optical interferometry, phase conjugate resonators, two wave mixing (beam couplings), and optical bistability. An improved method for testing the collimation of a laser beam was devised. The applications of a phase conjugate Twyman-

Green interferometer for testing lenses, concave and convex mirrors and coincidental surfaces such as paraboloidal mirrors were demonstrated. Different semilinear, linear and ring phase conjugate resonators were studied. By adjusting the feedback as well as the dimensions of any of the individual oscillators, any two of these resonators can be made bistable, i.e. if one resonator is pumping above its threshold level, the other should be below this level. Optical bistability phenomenon between different configurations of phase conjugate resonators was also studied. The signal in one phase conjugate resonator showed hysteresis as a function of the feedback into another resonator.

The third section of the report is concerned with research findings of Mr. Bo Su Chen, a current predoctoral student who is expecting to graduate in May of 1994. Mr. Chen has worked in several different areas of optical phase conjugation. Firstly he studied the Aureola electric-field superlattice around photorefractive holographic gratings. Secondly he investigated parallel image transmission via multimode Fiber-Photorefractive crystal system using phase conjugation. Thirdly, he has shown that reflection grating in [111] $\text{Bi}_{12}\text{TiO}_{20}$ crystal can be used for dynamic interferometry with low power He-Ne laser, giving diffraction efficiency 0.14%. The possibility of hologram recording by orthogonal polarized waves was also confirmed.

The fourth section of the report deals with the research findings of Mr. Y. Ding. Mr. Ding worked on two wave mixing. He explored a new configuration for mutual phase conjugation (MPC) using barium titanate crystal for nonlinear medium.

The fifth section of the report is concerned with the research work of Mr. Mohammad Mehdi who is currently a predoctoral student. Mr. Mehdi is investigating the energy upconversion phenomena. This technique allows one to

upconvert infrared radiation to the visible region enabling one to develop infrared detectors using conventional photomultiplier tubes for detection without cooling. Upconversion has also a host of applications in laser industry.

The sixth and the final section of the report presents the list of publications in scientific journals as well as the list of presentations made at various scientific conferences.

Section One

The nonlinear optical characteristics of organic materials were investigated by Dr. Hossin Ahamed Abdeldayem. He investigated degenerate four wave mixing in Disodium fluorescein doped Boric acid glass and measured the resonant as well as nonresonant third order susceptibility of 7'-dicynoapocarotene in Hexane. He studied the self action effect and the third order nonlinear susceptibility of "Chinse Tea" and the phenomenon of beam fanning in organic solutions.

Optical phase conjugation by degenerate four wave mixing has been an area of intense research, which has been studied in many organic and inorganic materials using pulsed as well as continuous wave lasers. Organic dyes doped in glasses and other solid materials have recently emerged as good optical phase conjugators. Fujiwara et al¹ examined the phase conjugate signal generated by degenerated four wave mixing (DFWM) and holographic process from a film of dried gelatin doped with fluorescein. They were able to recover a distorted image of an object by allowing the beam to pass twice through the distorting medium, after it was reflected from the fluorescein film. The decay time recorded in their experiment is of the order of 90 sec. The great need for nonlinear optical materials that can be used with low intensity light sources for the applications and the impressive results of fluorescein dye doped boric acid glass, obtained by Kramer et al² was the impetus to investigate the nonlinear optical properties of a different salt of the fluorescein family of compounds- Disodium Fluorescein in boric acid glass.

Disodium fluorescein has the chemical formula of $C_{20}H_{10}Na_2O_5 \cdot 2H_2O$, and its molecular weight is 412.31. Thin films of Disodium Fluorescein in boric acid glass were prepared using the technique developed by Todorov et al³. Six mixtures of boric acid and disodium fluorescein powder were prepared with concentrations ranging between 10^{-2} and 5×10^{-5} mole fraction of the dye in boric acid. Each mixture was heated in a glass tube almost $300^\circ C$ using a Bunsen burner for 15 to 20 minutes until the mixture became glassy and transparent. The molten mixture became a hard glass after a few seconds and small pieces could be taken from it and placed on a preheated glass slide at $120^\circ C$ and covered by a second hot slide. The two slides were then picked up and after a few seconds press hard to form the thin film.

An experiment was performed to record simultaneously, both the fluorescence and the phosphorescence spectra of Disodium Fluorescein in boric acid glass. The cw Ar^+ laser was chopped by a beam chopper. The signal from a photomultiplier tube is split into two signals, one of the beams is given to one gated integrator of the box car integrator to record the fluorescence spectrum, and the other is pre-amplified and fed to another integrator to the phosphorescence. These spectra were recorded using several laser lines.

The degenerate four wave mixing (DFWM) experiment was performed to obtain the phase conjugate signal. The phase conjugate reflectivity as a function of the pump beam intensity for different concentrations of the dye was measured, using the 488 nm line of the Ar^+ laser. The results have indicated that the reflectivity increases non linearly with the pump intensity. The high phase conjugate reflectivity obtained from the experiment lead to the examination of the possible mechanism responsible for the formation of the nonlinear grating. As the dye molecules are held rigid in the boric acid glass, the only possible mechanisms are the thermal effects and the population effects of

the singlet and triplet excited states. However, the other mechanism present in the solids, such as lattice vibrations, spin waves, acoustic waves, etc., usually contributes very little.

The possibility of the presence of the thermal grating in the sample was examined by changing the grating spacing from $0.6\mu\text{m}$ to $4.3\mu\text{m}$ by changing the angle between the probe beam and the writing pump beam (in DFWM experiment) from 114° to 12° . The phase conjugate signal decay time was measured by shutting off the probe and the writing beams simultaneously, allowing only the reading beam to fall on the sample. The two decay times from each decay curve for angle are 0.125 sec and 0.034 sec at the 114° angle, and 0.137 sec and 0.036 sec at 12° angle between the pump and the probe beams. These results reflect no change with the change in grating spacing. Consequently, the presence of the thermal grating was ruled out, where the thermal diffusion time should be different for the different spacing. The population grating could be due to either the singlet excited state or the triplet state or both. The fluorescence life time was measured to be of the order of a few nanoseconds (2 or 3 nsec). The result showed that the first triplet state is the one most responsible for the refractive index grating in the thin film samples used in our experiment. This conclusion was further confirmed by the measurements of grating and phosphorescence decay rates. The third order nonlinear susceptibility measurements indicated that the disodium fluorescein in the boric acid glass is a highly nonlinear material which shows a reasonable nonlinear response at a relatively very low power. It also shows a very high phase conjugate reflectivity which makes it perhaps the most efficient phase conjugate mirror available to date.

7'- dicyno-apocarotene⁴ is a relatively long molecule, whose mole weight equals 464.7, and has the molecular form $\text{C}_{33}\text{H}_{40}\text{N}_2$... The nonlinear

characterization of this sample was obtained by measuring the resonant enhanced third order nonlinear susceptibility as well as the non resonant one for different concentrations. The nonresonant third order nonlinear susceptibility was measured by the self-trapping technique, where the optical Kerr effect is the more dominant mechanism in this case. Self-trapping mechanism is a special case of self-focusing effects. Self-focusing, self-defocusing, temporal and spatial self-phase modulations, self-diffraction and beam fanning are all falling under the general name of self-action effects defined as those effects in which the nonlinear polarization induced by the incident beam affects the propagation or other properties of the same beam⁵. Self-focusing occurs as the result of a combination of a positive nonlinear index and a spatial variation of the laser intensity in which the beam is more intense in the center than the edges. In the presence of the optical field the increase in the refractive index follows the intensity profile of the incident beam, larger at the center and smaller at the edges. This profile corresponds to a lens and the beam focuses.

The resonant enhanced third order nonlinear susceptibility obtained by DFWM technique is of the order of 1 esu and that obtained by self-trapping technique, is of the order of 10^{-5} esu. It can be seen that the mechanism contributing to the DFWM experiment is very much different from that contributing to the self-trapping one. The laser line 532 nm in the DFWM is in resonance with the medium and at high power to pump enough number of molecules to the excited state and form a population grating. On contrary the cw Ar⁺ laser power is so low (~ 100 mw) which cannot by any means form similar population in the excited state. It is only contributing to the molecular orientation through Kerr effect which of course is much less efficient than the resonant effect.

The samples were prepared by soaking 4-Chinese green tea bags in ethanol for nearly 48 hours, and filtered to form a highly concentrated mother solution. The concentration was measured by evaporating a known volume of the original solution. A 10 mm rectangular cuvette was used to hold the sample. The absorption spectrum of Chinese tea showed a very broad absorption band ranging from UV to IR. The second harmonic of Nd:YAG laser is strongly absorbed in the solution and a red fluorescence is emitted. The fluorescence decay time was obtained to be a few nanoseconds (2 or 3 ns.) and it was shown to be pump beam polarization dependent, i.e. , the emitted red fluorescence is seen only in the direction perpendicular to the polarization direction of the exciting laser beam. The third order nonlinear susceptibility of Chinese tea in ethanol by degenerate four wave mixing, using a 532 nm from a pulsed Nd:YAG laser is attributed to resonance enhancement. The critical power for self trapping of a cw Ar⁺ laser at 514.5 nm, where the kerr-effect is the dominating mechanism gave $\chi^{(3)} = 10^{-5}$ esu. The multiple diffraction rings due to spatial self-phase modulation as well as other self action effects were also examined.

In one experiment we used the second harmonic of the mode-locked Nd:YAG laser and two orthogonally polarized beams, one above the other with nearly 2 mm separation are allowed to fall simultaneously on the solution. The P-polarized beam has an intensity of 40 W/cm², while the S-polarized beam has 10 W/cm² intensity. Once the laser is turned on, two sets of overlapping concentric rings could be observed within the first 0.01 second, after which the circularly symmetric pattern takes the form of half circularly symmetric pattern showing also that the weaker s-polarized beam remains circularly symmetric all the time. This multiple ring diffraction pattern is the result of laser induced refractive index change Δn in the solution. The response time of the molecular orientation is often of the order of a few picoseconds. When a linearly polarized

beam propagates through the tea solution the optical field induces dipole moments in the tea molecules which align themselves along the field. Furthermore the molecules spatially redistribute themselves in order to minimize the free energy of the system. Both molecular reorientation and redistribution lead to a change in the refractive index of the medium

Five different experiments were devised to measure the third order susceptibility of the tea solution in ethanol. These were 1-DFWM ; 2-Self-trapping; using the cw Ar⁺ laser; 3-Self-trapping technique using the mode locked Nd:YAG laser; 4-Spatial self phase modulation technique using cw Ar⁺ laser; 5-Spatial self phase modulation technique using the mode locked Nd:YAG laser. These five experiments revealed that the solution of Chinese tea in standard organic solvents gives rise to a rich variety of nonlinear optical phenomena. Large values of the third order nonlinear susceptibility ~ 1 esu from resonant enhancement and 10^{-5} esu from molecular orientation effects were obtained. The spatial self-phase modulation has also been demonstrated as a quick and simple technique to estimate $\chi^{(3)}$.

Beam fanning in solution is a new nonlinear phenomenon because it is different from the similar other nonlinear phenomena like beam bending or diffraction from a thin nonlinear grating. The beam bending phenomenon can be considered as a very special case of beam fanning, where a beam with nonuniform intensity distribution is bent with relatively small angle as it travels through a nonlinear medium, while in the beam fanning case, the beam fans within a wide angle. In the case of thin nonlinear grating the intensity distributions of different orders of diffraction behave in a similar manner as in the case of a linear grating, i.e. the lower orders of diffraction, near the beam axis, are more intense than the higher orders of diffraction, at the larger angles from the axis, while in the beam fanning case, this situation is quite opposite where

the diffracted light is most intense at higher orders (at larger angles), and decreases in intensity near the axis. Also, the formation mechanism of the thin nonlinear grating, which has been seen only in liquid crystals, is different from the grating responsible for beam fanning in organic solutions. The thin grating in liquid crystals is induced by the interference pattern of two coherent beams in the medium, while in the beam fanning case, the grating is induced by the spatial self-phase modulation pattern of a single beam.

Beam fanning in solutions is a consequence of an intensity-dependent refractive index which can be induced by several mechanism such as molecular orientation (Kerr effect) , electrostriction, molecular redistribution, liberation and electronic distortion. Some of its applications are the resonatorless optical bistability, radiation protection, and power limiting⁷.

The experimental arrangement for the observation of beam fanning in the solutions is the same as that of self-trapping discussed earlier. In the observation process it was noticed that the fanning of the beam is not stationary and shows fanning for 2 or 3 seconds and disappears. This process repeats itself. The threshold power for fanning is found to be dependent on the concentration of the solution. It is important to note that those radiation pressure forces which act as pulling and trapping forces for the atoms or molecules or a group of molecules are overcome by the thermal diffusion forces (Brownian force), which are several orders of magnitude larger. When a group of molecules are trapped at the focus and the fanning occurs, the thermal forces build up and diffuse them away from the focus field, and fanning disappears⁸.

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SECTION TWO

Applications of optical phase conjugation to beam couplings, resonators, bistability, and interferometry in electrically poled photorefractive BaTiO₃ crystal were investigated by Dr. Mostafa Dokhanian. This crystal is an efficient nonlinear material for optical phase conjugation with low power continuous wave lasers in the visible and near infrared. The light-induced change of refractive indices in electro-optic crystals are based on the spatial modulation of charges by nonuniform illumination. Electrons or holes are photoexcited from impurity centers present in the material and upon migration, are retraced at other locations, leaving behind them positive or negative charges of ionized trap centers. The photoexcited charges are then reexcited until they finally drift out of the illumination area. Therefore a space-charge field between the ionized donor centers and the trapped charges will be created which modulates the refractive indices via the electro-optic effect. Photorefractive effect was first discovered in 1966 by Ashkin¹ and then was utilized by Chen² for the storage of volume phase hologram. Huingard³ performed the optical phase conjugation in photorefractive materials using milliwatt laser beam. Feinberg⁴ showed that the photorefractive barium titanate can act as an efficient phase conjugator even with weak beams. Feinberg and Hellwarth⁵ then demonstrated the first cw phase conjugator having a reflectivity greater than unity by taking advantage of large Pockels coefficient of Barium titanate. They found that if a crystal is placed near a shiny mirror, this phase conjugator will track the reflective surface and direct a

beam of light normal to it. At the same time, Feinberg⁶ demonstrated a self-pumped phase conjugator consisting of a single photorefractive crystal.

Photorefractive effect is based on the transit of photoexcited carriers which can be affected by three different mechanisms: (1) diffusion, (2) drift, and (3) photovoltaic effect. However, in different materials, any one of the transport processes can dominate depending upon the electronic properties and experimental conditions. Barium titanate crystal was one of the first ferroelectric materials to be recognized as photorefractive. Its particular advantage for photorefractive applications is its large value of the electro-optic tensor component r_{42} which is responsible for large values of grating efficiency, beam coupling gain, and phase conjugate reflectivity. Such large reflectivity are particularly desirable in phase conjugate resonator applications, where to date BaTiO₃ is the material of choice. It is a member of the perovskite family of ABO₃ compounds. The Curie temperature of BaTiO₃ is 130° C. Above this temperature the crystal has a cubic structure with point group m3m. At the Curie temperature, it transforms to a tetragonal, ferroelectric phase with point group 4mm symmetry. This phase of the crystal is stable down to about 9°C at which temperature, a transition to an orthorhombic phase occurs. At -90°C a third and final transition to a rhombohedral phase belonging to group 3m symmetry takes place. The energy band structure of the crystal is determined by the titanium 3d orbitals which are responsible for low lying levels of conduction band and the oxygen 2p orbitals which are responsible for the upper levels of the valence band. The magnitude of its electro-optic coefficients are $r_{42}=820$ pm/V, $r_{13} = 8$ pm/V, $r_{33}= 28$ pm/v. The indices of reflection of the crystal at 30° C are $n_o = 2.437$ and $n_e = 2.365$ at $\lambda= 514.5$ nm. However one of the disadvantages of BaTiO₃ is its long response time typically of the order of 0.1-1 sec. for an intensity of 1Wcm⁻². The chemical reduction should lead to an improvement in the photorefractive

properties of as-grown crystal. When that sample is heated in a reducing atmosphere, the concentration of oxygen vacancies increases, and this results in a reduction of the valance state of the iron impurities, along with an increase in the population of free electrons.

Grating formation in photorefractive crystals is accompanied by an intensity redistribution between the interfering beams, i.e. the pump beam and a signal beam usually at a lower intensity. In a typical nonlinear medium, the self-interference of the incident beam with the diffracted beam creates a new holographic grating which can add to the initial one. Since the diffracted wave is phase delayed by $\pi/2$ with respect to the reading beam, the maximum energy transfer is obtained when the incident fringe pattern and the photoinduced index modulation are shifted by $\pi/2$ with respect to the reading beam, the maximum energy transfer is obtained when the incident fringe pattern and the photoinduced index modulation are shifted by $\pi/2$, thus corresponding to the spatial shift by $\Lambda/4$. In photorefractive crystals, such a $\pi/2$ phase shift exists when the recording is by diffusion of photocarriers. Consequently, a permanent and efficient amplification of a low intensity signal beam is possible. The maximum beam coupling in BaTiO_3 is obtained when the incident beams have the polarization parallel to the C-axis of the crystal. The large values of the gain coefficient in this crystal permits the amplification of a low intensity signal beam containing spatial information. For beam coupling and self pulsation's experiments we have used an Ar^+ laser at 488 nm in both multimode and single mode, and an electrically poled BaTiO_3 crystal to form different sets of resonators. The experimental configuration is shown in Fig. 1. The angle of incidence is not very critical. In one experiment the ring passive phase conjugator (RPPC) and the auxiliary resonators M_1C and M_2C are formed with multimode laser. It was found that the auxiliary resonator can be made to run in

a bistable or in coexisting mode by varying the relative reflectivity of the mirrors or by inserting suitable attenuation in the oscillator. It is also found that either of these two oscillators would continue to operate even after the ring is put off. Fig. 2 shows the recording these signals at the detectors D₁, D₂, and D₃ which measure the oscillations in RPPC, the combined phase conjugate signal, and the auxiliary resonator M₃C. One can see that if the signal increases in one, it simultaneously decreases in the other. With the laser in single-mode, for auxiliary resonators could be sustained simultaneously. Any two of them closing the other two, could be made to operate in a bistable condition, adjusting the relative reflectivities of the other mirrors. In another experiment, when any two of these oscillators were made to operate without RPPC, bistable oscillations were observed.

The phenomenon of bistability can occur only in nonlinear systems with a suitable feedback. A physical system is said to be bistable if in responding to one value of stationary input signal, its output could take two possible values which depend on the previous history of the system and on its fluctuations. In an optical system, nonlinearity alone is not sufficient to assure bistability; it is the feedback that permits the nonlinear transmission to be multivalued. In our experiment, we used a horizontally polarized Ar ion laser beam incident on an electrically poled BaTiO₃ crystal (8mm x 8mm x 8mm) in a face suitable for self-pumped phase conjugation. The orientation of the crystal with respect to the incident light is kept in such a way that no self-pumped phase conjugation is observable without either of the two semilinear resonators in operation. The light beam from the laser, after passing through the crystal, generates a broad fan of amplified light scattering toward the two external variable beam reflectors (attenuators) which are used to reflect the fanning light back into the crystal. This seeds bistable oscillations between the crystal and the reflectors. In a

semilinear resonator configuration, the variable beam reflector directs the beam onto an external mirror, which reflects the beam back to the crystal forming a ring resonator (Fig. 3). This results in the formation of bistable oscillations between the crystal and the semilinear resonator on one hand, and between the crystal and ring resonator on the other. Fig. 4 shows the bistability and hysteresis observed in the signal intensity of the ring resonator as the reflectivity of the semilinear resonator is first decreased and then increased in a cycle.

One primary application of optical phase conjugation is in the field of optical interferometry. we demonstrated the application of a phase conjugate interferometer For the testing of a concave spherical mirror and for measuring the refractive index of a liquid and an etalon. Fig. 5 shows the schematic diagram of a phase conjugate Twayman-Green interferometer for measuring the index of refraction of an etalon. In our experiment a parallel plate such as a Fabry-Perot etalon is placed perpendicular to the optical axis in the path of diverging beam of laser light. The collimated beam of light is obtained by focusing the laser beam at the focus of a well-corrected collimating lens by means of a microscope objective. The collimation is checked by observing straight and equidistant fringes on the screen. The collimating lens is mounted on the carriage of a precision distance device such as a differential micrometer with a least count of $1\text{ }\mu\text{m}$. The position of the lens is noted down on the scale of the carriage for the best collimation position of the lens as indicated by position 1 in the fig. 5. A parallel plate such as a Fabry-Perot etalon is now introduced between the microscope objective and the collimating lens. Therefore, the laser beam collimation is disturbed and the decollimation is shown in the shape of curved fringes in the interferometer. The collimating lens is then moved away from the microscope objective in order to obtain straight fringes again in the field

of view. The second position of the lens is noted as shown by position 2 of Fig. 5.

This is a nondestructive technique for the measurement of the index of refraction of Fabry-Perot etalon. The high reflecting multilayer dielectric coatings are not spoiled during the measuring procedure whereas the coating is spoiled in the liquid immersion technique for measuring the refractive index of optical components. When one the plane mirrors in the conventional Twayman-Green interferometer is replaced by a phase conjugate mirror, the interferometer becomes more sensitive for detecting a slight amount of laser beam collimation. It is possible to measure defocusing of the order of one micron for a lens of relative aperture $f/2$ in the phase conjugate Twayman-Green interferometer. However, the sensitivity for measuring the defocusing is reduced to about 8 microns for a collimating lens of relative aperture $f/5.7$. This uncertainty in the measurement of D results in the uncertainty of about 0.0004 in the refractive index measurement. The wave aberration introduced by the parallel plate of thickness 40 mm and refractive index of 1.5 when placed in the diverging beam of light for a relative aperture of $f/5.7$ has been calculated. The maximum optical path difference introduced by the parallel plate for the proper choice of the reference point is less than $\lambda/20$.

The phase conjugate interferometer offers several advantages for measuring wavefront distortions over other interferometers. The interferometer is free from aberrations due to self-focusing property of a phase conjugate mirror which is the BaTiO_3 crystal in our case. The interferometer does not require a good quality spherical mirror in its reference arm unlike Williams interferometer. It is capable of testing optical systems of smaller f -numbers. The interferometer can be used as a test plate for the testing the spherical surfaces. The accuracy

of the measurement of the refractive index of liquids or solid transparent material is 0.0004.

In another set of experiments we have used the phase conjugate Twyman-Green interferometer for examining the laser beam collimation which is shown in fig. 6. For increasing the aperture of the phase conjugate mirror, the expanded laser beam is focused on the barium titanate crystal by a positive lens. This lens needs not be of high quality because wavefront distortion introduced by the lens are compensated by the PCM. We used a 25 mW He-Ne laser along with a collimating lens of diameter 50 mm and focal length 250 mm, a focusing lens of diameter 50 mm and focal length 100 mm and a barium titanate crystal as phase conjugate mirror. When a defocused laser beam is incident on the beam splitter, circular fringes were obtained. In order to obtain a well collimated beam, the collimating lens was displaced longitudinally on the optical axis, until straight fringes were observed in the field of view. When the collimating lens is displaced slightly along the optical axis in order to obtain a slightly diverging beam of light, fringes of slight curvature are obtained. When the laser light is focused exactly on the focal point of the collimating lens, the fringes are straight. Hence this is most sensitive method for focusing the laser light on the focal point of the collimating lens and thereby obtaining a high degree of beam collimation. For measuring the beam divergence of the laser beam, the microscope objective, the spatial filter, and the collimating lens were removed from the set up of Fig. 6. The fringes of slight curvature were obtained to indicate a slight amount of beam divergence. Therefore, from this divergence of the fringes, the divergence of the laser beam was obtained using a suitable mathematical relation. We found the divergence of the beam to be 0.6×10^{-3} radian for the laser source in a good agreement of 0.66×10^{-3} radians given by the supplier of the laser.

In conclusion we have demonstrated a novel method for improving the laser beam collimation using the phase conjugate Twyman-Green interferometer. A theoretical relation was derived for detecting the minimum amount of the defocusing in the interferometer. It is possible to detect defocusing of the order of 1μ for a diffraction limited lens of f-number one. The technique has been found to be suitable for measuring the divergence of a laser beam.

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Fig. 1. Experimental set-up for observing phase conjugate resonators and bistabilities. B.S. - Beam Splitters, C-BaTiO₃ Crystal, M-mirror, D-Detectors, RPPC-Signal from Ring Passive Phase Conjugator.

Fig. 2 Signals from the detector D₁, D₂, and D₃ representing the ring oscillator (RPPC), semilinear oscillator M₃C, and the total phase conjugation using a multimode Ar⁺ laser. Ring is put off after 2.3 minutes. The RPPC and MC, are bistable. aa',bb', cc' represents a few points of bistability.

Fig. 3 - Experimental arrangement for bistability studies between a semilinear and a Ring Resonators. C: crystal, V.B.R.: Variable Beam Reflector, BS: Beam-Splitter, M: Mirror and D: Detector. M ,C,VBR #1 correspond to Resonator #1 and M ,C Resonator #2.

Fig. 4 — Bistability observed in the signal intensity of the ring resonator #1 versus the reflectivity of resonator #2.

Fig. 5 — Schematic diagram of a phase conjugate Twyman-Green interferometer for checking the laser beam collimation.

Fig. 6 — Schematic diagram of a phase conjugate Twyman-Green interferometer for measuring index of refraction of an etalon.

Section Three

This section of the progress report deals with the research work of Mr. Bo Su Chen; a current predoctoral student. He investigated the image transformation and real time interferometry in the fiber-photorefractive system using optical phase conjugation and its applications in optical fiber sensors as well as the reflection holographic grating in BTO crystal for real -time interferometry.

Image information by phase conjugation requires passing twice through the distortion. That is a major disadvantage in practical fields where the picture information is to be transmitted from one place to the other. We have demonstrated the abilities of performing optical processing using two-wave and four-wave mixing of one-way phase conjugation with the ferroelectric crystal BaTiO₃. We have studied holographic methods of one-way image transmission using photorefractive crystal in transmission and reflection geometries. Fig.1 shows a two-wave mixing schematic of holographic correction of a phase distortion. A phase distortion D is introduced in the signal wave A_S . Signal beam A_S and reference beam A_R are mutually coherent and they form an interference pattern inside a BaTiO₃ crystal. Due to photo-excitation and redistribution of the photocarriers, a volume phase grating is recorded with an index of refraction

amplitude $\Delta n \propto \frac{A_r A_s^*}{I_r + I_s}$, Where $I_r = |A_r|^2$ and $I_s = |A_s|^2$ are intensities of the signal and reference waves. Any reading beam A_s' is diffracted by the holographic grating. In Born approximation, for a small diffraction efficiency, the diffracted wave A_d is given by $A_d \propto \Delta n A_s' = \frac{A_r A_s^* A_s'}{I_r + I_s}$, When the reading beam A_s' has the similar phase distortion with A_s but different amplitude modulation, i.e. $A_s' = A_s T$, we have $A_d \propto \frac{A_r I_s T}{I_r + I_s}$. This means the diffracted beam contains the amplitude information but no phase distortion. This is a kind of holographic phase conjugation process through which one-way image transmission can be realized.

Fig.2 shows our experimental results. It is interesting to note that the dependence of grating amplitude on intensities of signal and reference beams in denominator, provides the advantage of performing optical processing with image correction at the same time. For instance, we can introduce image T_1 in the signal beam while writing the grating, and replace it by a different image T_2 during reading process. We will get an optical division in the diffracted wave (

for $I_s \gg I_r$): $A_d \propto \frac{A_r I_s T_2}{T_1}$. Similar results of image transmission with phase

conjugation were received in four-wave mixing with a reflection grating structure. In the Fig.3 configuration, beams A_1 and A_2 write a refractive index grating in a BaTiO₃ crystal. There is a distorting medium in the light path of beam A_2 . Since these two beams are incident from opposite sides of crystal, they create a reflection type grating. A third beam A_3 reads the grating in the count propagating direction of beam A_1 . Diffracted beam A_4 is the phase conjugate of beam A_2 which retraces the path of beam A_2 in the backward

direction. If we put a image T in beam A_1 , the holographic grating is proportional to $\sim A_1 T A_2^* e^{-i\phi}$. The diffracted beam can be written approximated by $\sim [A_1 T A_2^* e^{-i\phi}] A_3$. After that beam passes through the distortion, the phase distortion is canceled out and the information of the picture has been transmitted in one-way manner. A modified method is to put our image in the reading beam. In this case the grating will be as the form of $A_1 A_2^* e^{-i\phi}$ and the reading beam becomes $A_3 T$. The phase distortion also can be corrected. We have found that transmission gratings have broader spatial frequency bands, but reflection gratings are faster in time response.

In normal type of fiber interferometry, polarization-preserving single mode fibers are employed in both arms. Refractive index changes in one of the single mode fiber are induced by the field to be detected. This small index change integrates over the great length of the fiber (always many meters). The optical phase of the light passing through the signal fiber is thereby modulated. This phase modulation is then detected interferometrically, by comparing the phase of the light in the signal fiber to that of a " shielded " reference fiber. This approach is found to have some difficulties. Here we mention a few.

- Working with single mode fibers is rather difficult and expensive.
- Alignment is critical and not robust.
- Changes in refractive index of the single mode fiber are very small.
- The necessity of long lengths of fiber is inconvenient.
- Shielding one arm is complex.

We propose and demonstrate a real-time multimode fiber optic interferometric sensor. Instead of a polarization-preserving single mode fiber, we use a multimode fiber and a self-pumped phase conjugation mirror (PCM). Compared with single mode fiber optic sensor, this device is proved to be easier to work with, more sensitive and compact.

Simple analysis shows that multimode optic fibers do three things which appear to preclude their incorporation in interferometers.

First, multimode fibers produce modal dispersion, and different guided modes in the fiber have quite different light paths. There is no phase association among them. Second, the wavefronts of the light passing through multimode fibers show a speckle pattern. Third, even if we input a plane polarized light, the output light from even a short piece of multimode fiber is randomly polarized, and therefore, not useful for interferometry. The disadvantages of multimode fibers for interferometric sensors can be eliminated by combining it with an optical phase conjugation mirror. Some years ago Fisher and Sternklar¹ showed that phase conjugation back through a multimode fiber could allow interferometry. Kyuma² et al and Yariv³ et al helped to explain this by providing theory and experiment on modal dispersion of polarization and its recovery by phase conjugation. Later Volkov et al⁴ showed that wavefront and polarization can be recovered in a 2 meter multimode fiber using phase conjugation.

Suppose plane polarized light is coupled into a short length of multimode fiber. The output light is depolarized. If we insert a polarizer co-aligned with the input and phase conjugate the surviving light, the light will reenter the fiber and propagate "backward" through it. Because we have selected the complex wavefront which propagates forward with no net change in polarization, the phase conjugate wavefront propagates "backward" with no net change in polarization either. The phase of the incident light is also restored. Thus, the backward conjugation wave and reference wave can interfere and form an interference pattern. In this way, multimode fiber can be made to behave somewhat like a polarization-preserving single mode fiber.

Using a multimode fiber in interferometry with a phase conjugation mirror has the following advantages.

- Multimode mode fibers are much cheaper and easier to work with.
- Multimode fibers can tolerate much high optical power.
- In multimode fibers, mode structure changes would dominate over index of refractive changes. Because mode structures can be very sensitive to environment factors, we have greater sensitivity and therefore less need for long lengths of fiber.
- Short length of fiber means there is no need for long coherent lengths of laser, and then shielding one arm in interferometric sensor becomes easier.
- The backward signal beam from PCM is self-aligned and it adapts to changing conditions.

Employing optical phase conjugation in a fiber interferometric sensor allows the device to have the properties somewhat like that of optical novelty filter. The phase conjugator works in a dynamic manner. If the environmental change occurs slower than the response time of the phase conjugation mirror, the PCM has time to respond to the external change. The phase modulation in the signal arm will be adaptive and no slow change is detectable. On the other hand, if external field changes faster than the response time of the PCM, the beam emerging from the PCM will not be a plane wave. There will be instantaneous fringes moving at the output of the sensor. After the conjugator has had time to respond, the fringe motion stops.

Since different materials, geometry, and light intensity all profoundly affect response time of PCM, we can select response times over a wide range.

The schematic of the adaptive multimode optic fiber interferometric sensor is shown in Fig.4. It is a type of Michelson interferometer. A long coherence length He-Ne laser is used as a light source. The incident beam is horizontally polarized, and divided by a beamsplitter. The reflected part acts as a reference

wave. The transmitted part acts as a signal wave which is coupled into a multimode optic fiber (~ 35 cm, 130 μm diameter) using a 5x objective. A polarization beamsplitter selects the horizontally polarized portion of the beam from the fiber output to utilize the large electro-optic coefficient γ_{42} in the BaTiO_3 crystal. The surviving part is incident into the crystal. The beam inside the crystal self-bends⁵ to the crystal corner and is reflected internally by the crystal surface. Through a degenerate four wave mixing process, a phase conjugate wave is generated and it retraces the path of original incident beam. Since the whole process is self-aligning, it is called "self-pumped phase conjugation⁶."

After twice passing through the multimode fiber, the phase and polarization of the incident wave are restored. The signal beam and reference beam (reflected by a normal mirror) then interfere each other at the output of the device and form a clean interference pattern (Fig.5). Any external environmental change will modulate the phase of the signal light and causes the fringe motion. Conversation in the room is readily detected. Heat from a solder iron, inches away from the fiber produces dramatic pattern motion. Slight touching on the fiber causes the fringes move rapidly. The device is particularly sensitive to temperature change. When slowly inserting a human finger into the fiber loop (~ 6 cm diameter), we can see ~15 fringes motion in one direction. This is about a 5°C temperature difference, so we see three fringes/degree. We can measure to 1/100 of a fringe by heterodyne methods, so we have about 3×10^{-4} degree sensitivity with a single coil. N coils would give ($3/N$) 3×10^{-4} degree sensitivity. Taking back the finger, the fringes move backward again. Under the same conditions, no fringe motion can be observed when using a pen or other material. We are currently in the process of quantifying the parametric dependence of sensitivity.

We have discussed and showed that by using two wave mixing and four wave mixing with optical phase conjugation in BaTiO_3 photorefractive crystal, one-way image transmission can be realized with transmission and reflection geometries. And increased sensitivity of a multimode fiber allows us to achieve good sensitivity even with a short length fiber. Comparing with single mode fiber sensor, the adaptive multimode fiber optic interferometry with phase conjugation mirror is self-aligned, easier to work with, less expensive, compact and more sensitive to environmental change.

Here, we present the experimental investigation of reflection gratings in $\text{Bi}_{12}\text{TiO}_{20}$ (BTO) crystal. We use [111] cut crystal which provides for holographic grating vector $\mathbf{K} \parallel [111]$ isotropic reading, without introducing birefringence. We show the crystal natural optical activity, that usually hampered holographic recording in transmission grating⁷⁻⁹ can be used as an advantage that allow to write by orthogonal polarized waves.

The basic features of the dynamic holographic interferometer with reflection gratings is shown in Fig. 6. Two counter-propagating beams intersect in the crystal, beam a acts like an object and beam b as reference. The interference pattern is recorded in crystal via modulated crystal refractive index by electro-optic effect.

Beam "b" at the same time can participate in reading out in real time, producing beam a_d which may be different by polarization from transmitted object beam a_t . This polarization difference allows to select diffracted signal by analyzer. If some changes occur in the object beam, which results in small deviation of interference pattern, a new grating begins to form in the crystal and old grating is erasing. This leads to a simultaneous appearance of two diffracted beams from old and new holographic gratings. That generates interference fringes on the screen, which can be recorded by the video camera.(Fig. 7)

Besides being adaptive, DHI allows to work with "natural" unexpanded laser beams, which provided small enough objects to be tested by interferometry.

Another advantage of DHI, specific to optically active crystal BTO and reflection geometry is the possibility to write also grating with orthogonal polarized beams, which increase information ability of DHI.

To study the dynamic holographic interferometer of BTO crystal, we used the standard reflection grating (Fig. 6). A low power He-Ne laser used as the light source. In order to obtain large interaction region, two writing beams came from the same laser interacted inside the BTO crystal approximately at an angle of 160 degree. The intensity of both writing beam were kept at the level of 5 mw. They formed a interference patter. Through linear electro-optic effect, the refractive index of the crystal was modulated and generated a holographic grating. The crystal was oriented in such a way that the wave vector of the grating was directed along the direction of [111]. The writing beams acted also as the reading beams. They were diffracted by the grating in the reciprocal directions. Detector D and a HP 7090A plotter were used to measure the diffraction efficiency, grating buildup and decay time.

In our experiment, different polarizations of the writing beams were used in two different cases. In first case, both writing beams were parallel polarized. In second case, beam a was horizontally and beam b was vertically polarized. 0.14% diffraction efficiency for parallel and 0.11 % for orthogonal polarized waves were obtained from the reflection holographic gratings. For the first case, grating decay curve showed two-exponential erasure for diffraction beam ($\tau_1 = 0.55$ sec., $\tau_2 = 6.6$ sec.). That indicated existence of two mechanisms of grating information: fast and slow. Fast response can be described by photorefractive mechanism, but for slow response, we can only suggest about its possible

electrooptic nature. Fig. 8 shows the grating buildup and decay curves respectively.

In order to test the dynamic property of the system, we inserted a glass slide in the optical path of the beam a. When titling the slide, both diffracted beams from the old and the new gratings will appear simultaneously and interfere each other to form a dynamic interference fringes in a short period (this period depends on the time of the grating decay and buildup.). This conformed that our adaptive interferometer can be considered like a phase sensitive novelty filter which can visualize the movement of transparent objects. Since BTO crystal allows to write grating with orthogonal polarized waves, we used a darkening polarizer to block the transmitted beam and to select the diffracted beam. This will increase the signal to noise ratio.

Influence of the external electric field E_0 on beam propagation was demonstrated. We applied up to ± 3 kv/cm dc electric field along with the BTO [111] cut direction. The beam propagation direction also was in [111] direction. We observed large polarization rotation (30 degrees) and induced ellipticity of order 0.52 has been obtained. This implies BTO crystal has potential application in electro-optic modulation field.

To extend the application of this kind of reflection grating interferometer, we have tried to inject one of the writing beams through a short multi-mode optical fiber (core diameter $60\text{ }\mu\text{m}$). To reduce the laser power loss, the output of the fiber was not collimated and was directed very closely to the BTO crystal. Both writing beams formed a reflection grating. In our case, the depolarization of the optical fiber did not affect much on the grating and diffraction efficiency is around 0.12%, which can compare with our previous results.

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**Fig.1, Two-wave mixing schematic of transmission grating geometry.
D is a distortion and T is an Air Force resolution chart**

Fig. 2 To different transmitted images through a distortion medium using the experimental configuration of Fig. 1.

Fig.3, Four-wave mixing schematic of reflection grating geometry.

Fig.4, Schematic of a real-time multimode fiber optical interferometry with self-pumped phase conjugation mirror. M is a mirror, L_1 , L_2 , L_3 are lenses, P is a polarizer beam splitter and D is a detector.

Fig.5, Interference pattern of a multimode fiber optical interferometry. (center part has been over-exposed.)

Fig. 6, Reflection holographic grating in [111] cut BTO crystal. Beam a is an object beam, beam b as a reference beam. Beam at and ad are transmitted and diffracted beams respectively.

Figure 7 Dynamic holographic interferometric images "a" through "d", clockwise from upper left. The first figure shows the diffracted signal produced with no angular displacement in the image-bearing beam. The second figure shows the fringes observed on the diffracted signal when an x-y displacement is applied. The third figure shows the fringes observed on the diffracted signal when a horizontal or x displacement is applied. The last figure shows the fringes observed on the diffracted signal when a vertical or y displacement is applied.

Fig.8a the grating build up curve.

Fig. 8b, the grating decay curve.

Section Four

This part of the report is concerned with the research findings of Mr. Yao Ding, who worked on two wave mixing phenomena. He devised a new configuration for Mutual optical phase conjugation in a single domain BaTiO₃ crystal. In his experiment, two extraordinary beams 1 and 2, whether coherent or incoherent, are incident on a poled BaTiO₃ crystal on its positive C face at points P and Q, to cross at point R on the negative C face, generate mutually pumped phase conjugate beams 1* and 2* detected by D₁ and D₂ respectively (Figs. 1 and 2). The beams do not self pump individually. When the gratings are formed with two coherent beams 1 and 2, and if beam 1 is put off, the signal D₂ dies immediately while the signal D₁ goes up a little for about a second, and then decays in about 58 seconds. Here, $I_1 = I_2 = 1.5\text{mw}$ (Fig. 3). On the other hand, if beams 1 and 2 are put off simultaneously, and an incoherent beam 3 ($I_3 = 0.5\text{mw}$) is put on in the place of beam 2, signal D₂ dies immediately while D₁ goes up for the first 19 seconds and then decays in 2.8 minutes (Fig. 4), the growth and decay times decreasing with the intensity of beam 3 (or 2).

These observations indicate the formation of bragg grating at R responsible for the beam reflection in opposite channels, and reflection gratings along path PR and QR where beams are partially reflected backward. Thus when beam 1 is put off beam 3 (or 2) goes to detector D₁ which decays with grating at R. The effective signal at D₁ increases initially (Figs. 3, 4) because of the decay of the reflection grating along PR.

Figure 1. Experimental arrangement for two wave mixing studies. B.S.-Beam Splitter, D- Detector, M-Mirror, S- Shutter.

Figure 2. Enlarged version of a part of the two-wave mixing experiment: C-BaTiO₃ crystal, BS - Beam Splitters, 1 & 2, the laser beams, 1* and 2* , the phase conjugated beams

FIG. 3

Figure 3. After the phase conjugate signals 1* and 2* stabilize, beam 1 is put off. 2* decays abruptly while 1* initially rises for about a second and then decays in about 58 seconds. Because 1 and 2 are coherent, $I_1 = I_2 = 1.5 \text{ mw}$.

FIG 4

RANGES:	100.0V	1.000V	600.0mV
OFFSETS:	0.0V	0.0V	0.0V
TOTAL TIME:	4.00M		
POST-TRIG:	0.0S		
TRIGGER:	MAN		
00:40:17	14 FEB 92		

Figure 4. After grating formed by the coherent beams 1 and 2 are stabilized, beam 2 is put off and beam 3 which is incoherent to beam 1 is put on. The signal at D₂ decays abruptly while the line at D₁ grows for 19 seconds, and then again decays in 19 minutes. The shape at the maximum of the curve is spurious.

FIG. 5

RANGES:	100.0V	1.000V	600.0mV
OFFSETS:	0.0V	0.0V	0.0V
TOTAL TIME:	2.00M		
POST-TRIG:	0.0S		
TRIGGER:	MAN		
23:43:01 13 FEB 92			

Figure 5. This figure is similar to figure 3 except that $I_3 = 5\text{mw}$, because of which signal 1* at D₂ decays faster.

Section Five

This section of the report deals with the research findings of Mr. Muhammad Mehdi, a predoctoral student working on the concept of "energy upconversion". Energy upconversion in doped materials is a phenomenon in which one ion is excited to an energy level at ν_1 exchange energy with another ion at energy ν_2 in such a way that the second ion goes to its ground state while the other atom goes to higher energy level at $\nu_1 + \nu_2$ and fluoresces at energies greater than ν_1 . This method allows one to upconvert infrared radiation to the visible region enabling one to develop infrared detectors using conventional photomultiplier tube for detection without refrigeration. Using proper optical resonator configuration one can make use of the upconverted fluoresce to develop upconversion lasers (R. M. MacFarlane, Optics and photonics News Volume 3, No. 3, P. 8 (1992)).

In the present work Mr. Mehdi has recorded infrared upconverted fluorescence in $\text{LaF}_3 : \text{Nd}^{3+}$ and $\text{BaF}_2 : \text{Nd}^{3+}$. Near infrared laser beam at about 800 nm from a Ti:Sapphire laser excited by Ar^+ laser has been used to irradiate the single crystals. The upconverted infrared fluorescence in these two crystals are shown in figs. 1 and 2, and the wavelength of the individual peak are marked. The analysis is in progress. One can see that in $\text{LaF}_3 : \text{Nd}^{3+}$ at 657 and 589 nm are sharp and intense and they are very likely suitable for laser action. Similarly in $\text{BaF}_2 : \text{Nd}^{3+}$ the fluorescence at 662, 582 and 524 nm are suitable for laser action.

Fig. 1 UP CONVERTED FLOURESCENCE SPECTRUM OF BaF:Nd³⁺
EXCITED BY Ti:SAPHIRE LASER AT 806nm

Fig. 2 UPCONVERTED FLOURESCENCE SPECTRUM OF $\text{LaF}_3:\text{Nd}^{3+}$
EXCITED BY Ti:SAPPHIRE LASER AT 306nm

Section six

List of Publications and presentations

1. Transient multiple diffraction rings induced by ultrafast laser from chinese tea
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12. Phase Conjugate Resonators and Bistable Oscillations in BaTiO₃ Photorefractive Materials, Effects and Devices, OSA Jan. 1990
13. Phase Conjugate Resonators and Bistabilities in Photorefractive BaTiO₃, OSA Annual Meeting, Technical Digest, Vol. 16, P. 258 (1991)
14. New Configuration for Mutual Phase Conjugation in BaTiO₃, OSA Annual Meeting, Technical Digest, Vol. 23, P.178 (1992)
15. Reflection Holographic Gratings in {111} cut Bi₁₂TiO₂₀ for real time holography, accepted for publication by *Optics Communications*.
16. Optical Instability and Hysteresis With a Photorefractive Self-Pumped Ring Phase Conjugate Resonator., OSA Annual Meeting, Technical Digest, Vol. 23, P. 109 (1992)
17. Aureole Electric-Field Superlattice around Photorefractive Holographic grating, OSA Annual Meeting, Technical Digest, Vol. 16, P. 23 (1993)
18. Parallel Image Transmission by Means of a Multi-Mode Fiber-Photorefractive Crystal System Using Phase Conjugation, OSA Annual Meeting, Technical Digest, Vol. 16, P. 238 (1993)
19. One-Way Image Transmission via Media and Real-Time Interferometry in Photorefractive Crystal, SPIE 1993
20. Photorefractive Reflection Gratings in {111} cut Bi₁₂TiO₂₀ for adoptive Interferometry, OSA Annual Meeting, Technical Digest, Vol. 16 P. 59 (1993)

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