REPORT DOCUMENT PAGE

AFRL-SR-BL-TR-00-

0013

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
15 December 1999

3. REPORT TYPE AND DATES COVERED
Final technical report
15 Mar. 1997 through 14 Mar. 1998

4. TITLE AND SUBTITLE
Study of Photorefractive Effects in Periodically-Poled Lithium Niobate

5. FUNDING NUMBERS
Grant F49620-97-1-0221

6. AUTHORS
A. Alexandrovski, R. K. Route, M. M. Fejer

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESSES
Edward L. Ginzton Laboratory
Stanford University
Stanford, CA 94305-4085

8. PERFORMING ORGANIZATION REPORT NUMBER
SPO No. 17602

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AFOSR
801 North Randolph Street, Room 732
Arlington, VA 22203-1977

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES
The view, opinions and/or findings contained herein are those of the author(s) and should not be construed as necessarily representing the official policies or endorsements, either expressed or implied, of the Air Force Office of Scientific Research or the U.S. Government.

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT ***(Maximum 200 words)***

The objective of this DURIP-97 program was to facilitate investigation of photo-refractive effects in periodically poled lithium niobate (PPLN). Performance limitations due to photo-refractivity were studied using photo-thermal common-path interferometry (PCI) to determine if photo-refractive damage sensitivity is dependent on grating duty cycle. The PCI technique operates in a "pump-probe" configuration, and is sensitive to both linear optical absorption at the ppm/cm level and photo-refraction. Bulk and waveguide frequency doubling PPLN modules having a range of grating pitches were fabricated by Gemfire Corporation. In the bulk PPLN devices, optimum performance was found in modules with a nominal duty cycle close to the 50% value predicted by assuming that photo-refraction in lithium niobate is caused by photo-galvanic charge separation and that the alternating ferroelectric domain polarity due to the grating acts to "short-circuit" any charge build-up. Similar optimum performance is expected from waveguide harmonic conversion devices which have 40-50% duty cycles.

20000120 095

14. SUBJECT TERMS
Periodically-poled lithium niobate, PPLN, lithium niobate photorefractive effect, photorefractivity, periodic poling

15. NUMBER OF PAGES
15

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	**18. SECURITY CLASSIFICATION OF THIS PAGE**	**19. SECURITY CLASSIFICATION OF ABSTRACT**	**20. LIMITATION OF ABSTRACT**
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

Study of Photorefractive Effects in Periodically-Poled Lithium Niobate

Final Technical Report
for the period
15 March 1997 through 14 March 1998

Principal Investigator
Professor Martin M. Fejer
Applied Physics Department
and
Center for Nonlinear Optical Materials
Stanford University
Stanford, California 94305-4085

E. L. Ginzton Laboratory
SPO No. 17602

Report date
November 1999

Prepared for
AFOSR

Grant Number F49620-97-1-0221

TABLE OF CONTENTS

I. Table of Contents i

II. List of Figures and Tables ii

III. Technical Report 1

- A. Statement of the Problem Studied 1
- B. Background on the PCI Technique 1
- C. Theory of Photorefraction in Periodically-Poled Ferroelectrics 4
- D. Experimental Procedures 5
- E. Results 6
- F. Discussion 8
- G. Conclusions and Recommendations for Further Study 10

IV. References 10

V. Appendix - List of Equipment Purchased 11

II. LIST OF FIGURES AND TABLES

Fig.1: Crossed-beam setup for low absorption measurements 2

Fig. 2: Schematic of pump-beam effect 3

Fig. 3: GRIIRA in PPLN at 200°C, extraordinary beam 3

Fig.4: Crossed-beam geometry of the PCI technique used in this study 5

Fig.5: Illustration of a typical sample 6

Fig.6: Time-dependence of the detected probe beam power 7

Table1: Rate of the relative intensity change at the probe beam center 7

Table 2: Measurement of $\Delta I/I$ in saturation 8

Table 3: Steady-state index changes 8

III. TECHNICAL REPORT

A. Statement of the Problem Studied

The objective of this DURIP-97 University Research Instrumentation Program, F49620-97-1-0221, was to facilitate an investigation of the phototorefractive effect in periodically poled lithium niobate (PPLN). PPLN is a very important new nonlinear optical material with a large number of defense-related applications involving nonlinear optical frequency conversion. While it is well-known that bulk lithium niobate suffers from photorefractive damage, particularly in the green, the effect in periodically-poled structures is less well-known. We proposed, therefore, to determine the performance limitations imposed by photorefractivity on periodically-poled devices and to determine if its damage sensitivity can be predicted by a simple model that takes into account the geometric parameters of the photorefractive grating. We planned to characterize the behavior of commercially-fabricated PPLN bulk and waveguide frequency doubling modules through a series of highly sensitive "pump-probe" experiments, focusing on the severity of photorefractivity as a function of the nominal duty cycle of the (periodic) ferroelectric grating.

Custom-built bulk and waveguide PPLN frequency conversion modules having a range of grating pitches and duty cycles were fabricated by Gemfire Corporation, then known as Deacon Research. (Gemfire is a leading commercial manufacturer of PPLN devices with grating pitches and duty cycles in the range we were interested in for the proposed studies.) The bulk modules were designed for SHG of 1064 nm radiation and the waveguide modules for SHG of 780 nm and 960 nm radiation. In the study, we focused on the bulk modules since they were easier to measure and the data was straightforward to interpret. Measurements of index variations as a function of duty cycle were carried out in Stanford's CNOM Central Characterization Facility using photo-thermal common-path interferometry (PCI), a technique which is highly sensitive, both spatially and temporally, to very small optical distortions caused by the absorption of high intensity laser beams. In addition to the frequency conversion PPLN modules mentioned, capital items purchased during the program consisted of optical sources and analytical instruments useful for characterizing the PPLN devices.

B. Background on the PCI Technique

We typically use photo-thermal common-path interferometry (PCI) as a sensitive tool for the measurement of very low levels of light absorption in solids. Since it accomplishes this by detecting small thermally-induced changes in the optical index of refraction, it is also ideally suited for detecting photorefraction and perhaps elucidating the photorefractive damage mechanism as well. The PCI technique was developed originally for use in the chemical analysis of isotropic gases and liquids [1]. Numerous modifications of photo-thermal techniques all rely on heating of the sample by a pump beam, while a change in one or more physical properties of the sample caused by the heating are detected using a variety of methods.

In previous work, we have shown that the optimized near-field detection scheme developed at Stanford is fully as sensitive as interferometric methods. Therefore, we refer to it as 'common-path photo-thermal interferometry' (PCI). Simple analytical solutions have been developed for the temporal response of a dual-beam device, both in collinear and crossed-beam configurations. Chopped CW pump laser sources combined with lock-in detection provide very high amplitude and phase detection sensitivity. The frequency response of the PCI technique has been analyzed thoroughly, including the phase of the detected signal, its longitudinal and radial dependencies, the effect of finite detector aperture, signal-to-noise ratio, and optimization guidelines. A reliable and versatile crossed-beam setup that we have developed for measurement of low optical absorption in bulk samples is illustrated in Fig.1.

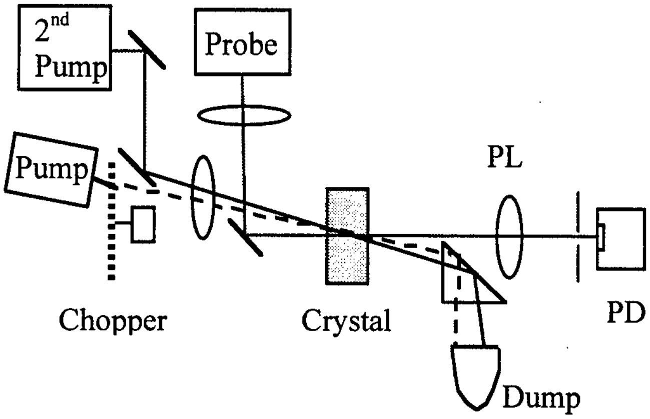

Fig.1: Crossed-beam setup for low absorption measurement: PL: projecting lens, PD: photodetector. A second pump beam is added whenever the influence of light of a different wavelength on the absorption of the pump beam is to be studied.

The interferometric sensitivity of the PCI technique implies that phase distortion $\Delta\phi$ of the probe beam due to the pump beam heating effect (see Fig.2) can equally well reveal corresponding intensity perturbations of the probe, $\Delta I/I \approx \Delta\phi$. For materials with dn/dT around 10^{-5}/K and with a pump power of 1W, this gives resolution of 10^{-6} (1 ppm) in terms of the absorbed fraction of pump power.

The other key feature of the PCI technique is its spatial resolution which corresponds in the transverse direction to the pump spot size. Its longitudinal resolution is equal to the effective interaction length of the crossed beams which can be reduced by increasing the

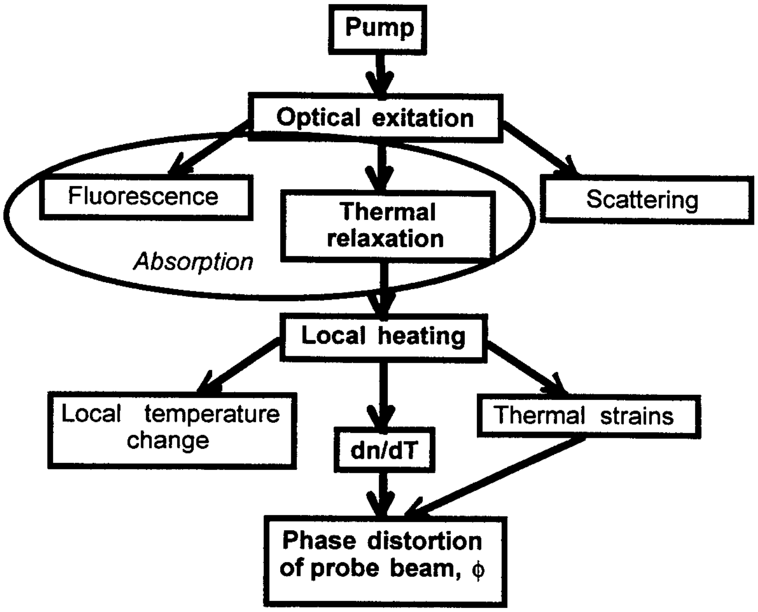

Fig. 2: Scheme of pump beam effect.

crossing angle. Spatial resolutions of 0.1x0.1x0.5 mm for the crossed-beam PCI technique have been demonstrated in studies on high-quality KTP crystals. The high sensitivity of the technique can easily be converted to a high temporal resolution. In Fig.3, the response of a periodically -poled lithium niobate (PPLN) crystal illustrates green-light-induced infrared absorption (GRIIRA), with temporal resolution of 30 ms. In this case, one pump beam (1W, 1064 nm) was focused into 50mm-long PPLN crystal along with the second pump beam (532 nm, 0.5 W) which was shuttered manually. A factor of three increase in the IR absorption is seen in the presence of the green light.

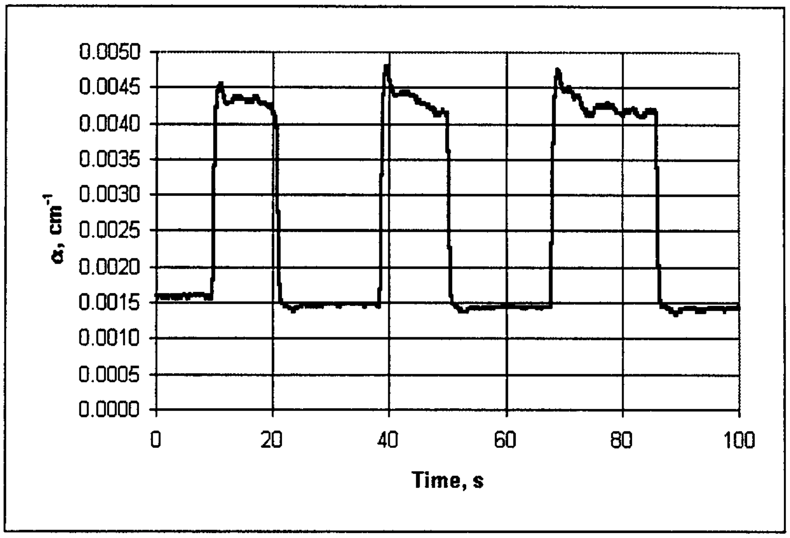

Fig. 3: GRIIRA in PPLN at 200°C, extraordinary beam.

With pump powers in the range 10mW-10W in a pump beam approximately 100 μm in diameter, it is easy to observe a variety of photo-chromic effects in optical materials: light-induced coloration, self-bleaching, bleaching by probe beam, etc. which are all phenomena seen in practical laser systems. This is the result of the PCI pump power densities being quite high compared to those used in commercial spectrophotometers.

C. Theory of Photorefraction in Periodically-Poled Ferroelectrics

A quantitative analysis of photorefractive effects in periodically-poled ferroelectrics has been published previously [3]. This analysis shows that photorefraction should almost disappear in a perfectly fabricated, periodically-poled material with 50% duty cycle due to 'short-circuiting' of the photo-galvanic effect. In realistic situations, there are at least three contributions to residual photorefraction in periodically-poled samples:

- effects due to the grating period being finite compared to the pump beam spot size [3],
- systematic deviations in the duty cycle of the periodic domain pattern from the optimum 50% value, and
- local errors in patterning, especially in the duty cycle.

From [3], one can derive relatively simple relationships to estimate the magnitude of these effects. In an 'ideal' case of a 50%-duty cycle with no patterning errors, the effect of a finite ratio of grating period to pump beam diameter can be estimated at the beam center by the following equation:

$$\Delta n_o \approx \frac{32}{\pi^2}\frac{1}{(K_g w)^2}\Delta n_h \qquad (1)$$

where Δn_0 is the index change at the center of the pump beam, Δn_h is the index change that would occur in a bulk sample with the same magnitude of the photogalvanic effect, K_g is the grating wave vector ($K_g = 2\pi/\Lambda$, where Λ is the grating period) and w is the pump waist radius.

The effects of systematic deviations in the duty cycle from the 'ideal' 50% can be estimated as well by equation (2):

$$\Delta n_0 \approx a_0^2 \Delta n_h \qquad (2)$$

where a_0 is the 'unipolarity' of the sample which can be defined here as $a_0 = 2d - 1$ (d is the duty cycle).

It is not as easy to calculate the contribution of the local errors in a domain pattern. Generally, if these errors create local unipolarity, on a scale longer than w, then expression (2) holds. Irrespective of which domain polarity dominates locally, the sign of the effect will be the same so that the photorefraction effect will accumulate along the pump beam path. In this way, expression (2) can be used with some average value of the patterning error a_0^2.

D. Experimental Procedures

The experimental setup is illustrated in Fig. 4 below. An extraordinary polarized CW green beam (514 nm) was focused into the sample to a waist radius of 35 microns. A red HeNe-laser probe beam was focused to a waist radius of 100 microns to probe the index change induced by the green pump.. The relatively large index changes (up to 10^{-3} for a single-domain sample) that occurred allowed the use of a relatively large crossing angle of 15° (in air) and therefore good spatial resolution due to the small value of L_{eff} in the medium.

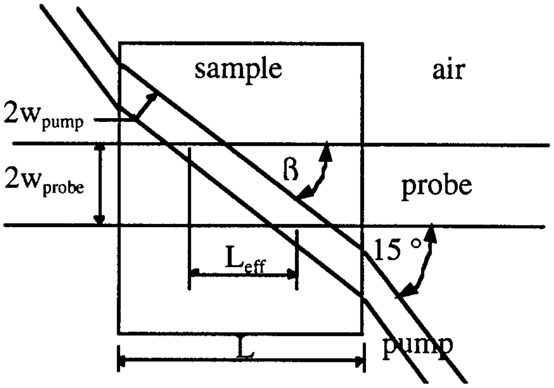

Fig.4: Crossed-beam geometry with beams intersecting at an angle β inside the sample. Intersection angles are exaggerated for clarity.

L_{eff} is given by:

$$L_{eff} = \sqrt{\frac{\pi}{2}}\frac{w}{\sin\beta}n \qquad (3)$$

where β is the crossing angle inside the crystal and n is the refractive index. A change in the optical index of refraction, Δn, causes a phase distortion of the probe beam given by:

$$\Delta\varphi = 2\pi\Delta n L_{eff} / \lambda \qquad (4)$$

This in turn results in an amplitude distortion (relative intensity change at the probe beam center) which reaches a maximum of approximately

$$\frac{\Delta I}{I} = \frac{1}{\sqrt{2}}\Delta\varphi \qquad (5)$$

at a distance of $z = 2.72\ w^2/\lambda$, where λ is the probe wavelength. It is this amplitude distortion that is detected with interferometric sensitivity by the experimental apparatus. (A manuscript on a complete analysis of the PCI technique is in preparation.)

Empirically, it was determined that the initial slope of the time dependence $\Delta I(t)$ correlated closely with its t → ∞ saturation value. By focusing on the initial slope and using a simple linear relationship to calculate the saturation value, we were able to avoid uncertainties in measuring photorefraction under saturation conditions.

Five bulk PPLN samples 10x20x0.5mm (XxYxZ), with grating periods of 6.0, 6.2, 6.3, 6.4, and 6.5 microns were studied. Each sample had three sections with photo-lithographic duty cycles of 40%, 50%, and 70%, respectively. The samples also had narrow unpoled end-sections (0% duty cycle) on either side of the periodically-poled sections as illustrated in Fig. 5 below. These unpoled sections were used to determine photorefraction for the homogeneous case.

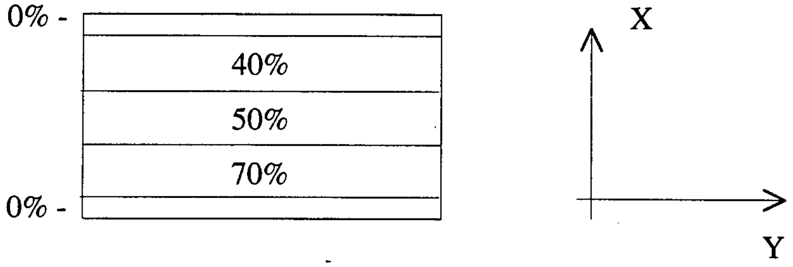

Fig.5: Illustration of a typical sample

E. Results

An example of the recorded time dependence of the probe beam intensity at the probe beam center is given in Fig. 6.

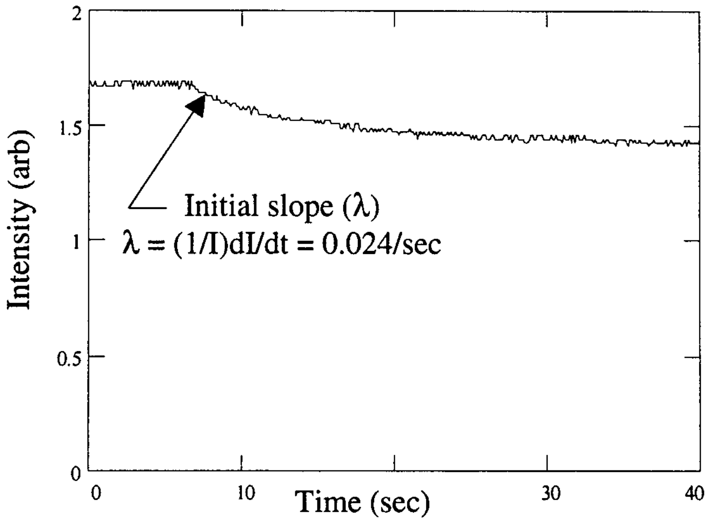

Fig.6: The time-dependence of the detected probe beam power: PPLN sample with the grating period of 6.4 microns, 50% duty cycle section.

Values for the rate of the relative intensity change at the probe beam center, **(1/*I*)*dI/dt***, are given in Table 1. The power density of the extraordinary polarized green pump was 800W/cm^2 in the periodically-poled regions. This was reduced to 100W/cm^2 in the unpoled sections where photorefraction was much higher. Several points in each poled section were probed with the pump/probe intersection near the front face of the sample.

Table 1: Rate of relative intensity change, **(1/*I*)*dI/dt*** (sec^{-1})

period (μm)	nominal duty cycle - 40%	nominal duty cycle - 50%	nominal duty cycle - 70%	unpoled region
6.0	0.006 0.006	0.25 0.055 0.12	0.15 0.25	0.05 (0.40)*
6.2	0.004 0.004	0.007 0.002 0.001	0.008 0.024	0.04 (0.32)*
6.3	0.001 0.0035	0.043 0.024 0.030	0.20 0.18	0.045 (0.36)*
6.4	0.004 0.005	0.055 0.024 0.028	0.10 0.09	0.12 (0.96)*
6.5	0.013 0.010	0.022 0.066 0.022	0.70 0.50	0.06 (0.48)*

()* data normalized to account for 8X reduction in pump power in unpoled regions.

The measurement of **Δ*I*/*I*** in saturation is given in Table 2, for selected points. Generally speaking, these data follow the data in Table 1, the corresponding time constant being 6-7 sec. Estimates of the corresponding change in extraordinary index are given in brackets.

Table 2: **Δ*I*/*I*** in saturation

period	nominal duty cycle - 40%	nominal duty cycle - 50%	nominal duty cycle - 70%	unpoled region
6.0				
6.2				
6.3				
6.4	0.029 (1.1×10^{-5})*	0.155 (5.9×10^{-5})*		
6.5	0.087 (3.4×10^{-5})* 0.096 (3.7×10^{-5})*	0.13 (5.0×10^{-5})*		

()* estimated extraordinary index changes in saturation. Equations (4) and (5) were used, with L_{eff} = 0.37 mm (inside the sample).

F. Discussion

Assuming that the saturation level **Δ*I*/*I*** is proportional to the slope **(1/*I*)d*I*/d*t*** for every measured value in Table 1, the steady-state index changes were calculated (Table 3) .

Table 3: Steady-state index changes,Δn_e x 10^5

period (μm)	nominal duty cycle - 40%	nominal duty cycle - 50%	nominal duty cycle - 70%	unpoled region
6.0	1.5 1.5	62 14 30	37 62	99
6.2	1.0 1.0	1.7 0.50 0.25	2.0 5.9	79
6.3	0.25 0.85	10 5.9 7.4	49 44	89
6.4	1.0 1.2	14 5.9 6.9	25 22	240
6.5	3.2 2.5	5.4 16 5.4	170 120	120

Except for the sample with a period of 6.2 microns, the lowest level of photorefraction was observed in the section with a nominal lithographic duty cycle of 40%. This observation suggests that these samples were slightly 'over-poled,' meaning that the regions with inverted polarity were slightly wider than the nominal lithographic grating features used to create them. The effective duty cycle of a ferroelectric grating is governed by the duty cycle of the lithographic grating, the poling voltage waveform, and the domain wall velocity / field characteristics, and is difficult to control with great precision. Being somewhat 'over-poled' would account for an increase in the effective duty cycles of the devices and cause the minimum in photorefraction to occur at an apparently lower nominal duty cycle than at 50% where it should theoretically occur. Sectioning, polishing and chemical etching of the PPLN gratings is the most unambiguous way to determine the effective duty cycle, but this is not particularly practical because of its destructive nature.

For the sample with the largest period, 6.5 microns, the photorefraction measured in the 70% region was equal to, or even stronger than that in an unpoled section. This was not interpreted as indicating that the duty cycle in this region was close to 100%. In previous (unpublished) work, we observed several instances where the photogalvanic effect appeared to be enhanced by the poling procedure, but this effect has not yet been studied or quantified.

Practically speaking, the results given in Table 3 demonstrate that photorefraction in the periodically-poled sample can be suppressed by more than two orders of magnitude. The 'floor' for the photorefraction in periodically-poled sample is given by eqn. (1) which under our experimental conditions is equivalent to a 'suppression' factor of 0.0026. Therefore, the "best" sections were probably near this 'floor'.

On the other hand, the suppression of photorefraction by fine pitch gratings is sensitive to duty cycle and a variance of only ±5% from the theoretical optimum of 50% will limit the suppression factor to a level of 0.01, as indicated by equation (2). This was roughly the case for the 40% sections of the samples with the shortest (6.0 microns) and longest (6.5 microns) periods.

G. Conclusions and Recommendations for Further Study

The major findings of this study include the following observations:

- A suppression of photorefraction in PPLN at a level between two and three orders of magnitude was observed.
- The theoretical limit of the residual photorefraction was probably reached, given evidence from other experiments for an enhanced photogalvanic effect in poled samples.
- The occurrence of a minimum in the photorefraction in sections with nominal lithographic duty cycle of 40%, as opposed to 50 % as predicted by theory, was interpreted as a degree of 'overpoling' in these samples.

These results show clearly that periodically-poled lithium niobate (PPLN) displays several orders of magnitude less photorefraction than bulk lithium niobate because its fine-pitch anti-parallel ferroelectric grating results in short-circuiting of the photogalvanic currents that would otherwise cause charge build-up and detrimental electro-optic index distortion. This should make it possible to operate PPLN at lower temperatures and/or higher power levels than is possible with bulk lithium niobate. However, the photogalvanic effect is an intrinsic property of the material and it cannot be eliminated entirely. Further reductions in photorefraction sufficient to operate PPLN devices at room temperature are estimated to require at least several more orders of magnitude improvement, which is challenging given current materials technology.

While only minor improvements are envisioned in PPLN devices, moving to nonlinear optical materials with intrinsically lower photorefraction such as lithium tantalate, MgO-doped lithium niobate, etc. seems like a more effective approach to achieving higher power, lower temperature device operation. Toward this end, we have initiated a collaboration with Crystal Technology Inc., Lightwave Electronics, and Silicon Light Machines to develop a process for fabricating and testing periodically-poled lithium tantalate devices. The improvements resulting from periodic poling will be multiplicative with respect to improvements in bulk material properties, so that it is reasonable to expect much lower levels of photorefraction in periodically-poled devices made from these newer materials.

IV. REFERENCES:

[1] S.E.Bialkowski, *Photothermal Spectroscopy Methods for Chemical analysis*, Chemical analysis series, **v.134** (John Wiley & Sons, New York, 1996).

[2] M.A.Olmstead, N.M.Amer, S.K.Kohn, D.Fournier, and A.C.Boccara, *Appl. Opt.* **20**, 1333 (1981).

[3] M. Taya, M. C. Bashaw and M. M. Fejer, "Photorefractive Effects in Periodically-Poled Ferroelectrics," *Optics Letters* **21**, 857 (1996).

V. APPENDICES

A. List of Equipment Purchased

PROPERTY REPORT FOR CONTRACT F49620-97-1-0221

Tag #	Dept	Description	Manufacturer	Model	Serial	Acq Date	TV	PO Num	Fund#	Account#	Acct Amoun
9118850	WMZ01	AMPLIFIER	STANFORD RESEARC	SR830	36045	25-Jul-97	1	G41066	169J265	2WMA521	4,018.81
9119116	WMZ01	CONTROLLER, CH	NEW FOCUS, INC.	3501	1017	26-Aug-97	1	G40292	169J265	2WMA521	1,623.75
9119741	WMZ01	COMPUTER	HIQ	RCZFO	9801140W/0737	8-Jan-98	1	G66056	169J265	2WMA521	1,303.87
9119932	WMZ01	COMPUTER	HIQ/E.C.W. ENTERPR	M9EHHO	9802149 W/O 7	11-Feb-98	1	G79813	169J265	2WMA521	1,769.89
9119949	WMZ01	ANALYZER, SPECT	ANDO ELECTRIC CO.,	AQ6315A	80639304	26-Feb-98	1	G77461	169J265	2WMA521	36,745.46
9282681	WMZ01	MODULE	GEMFIRE CORP.	NONE	NONE	1-May-98	1	G44196	169J265	2WMA521	12,991.02
9282698	WMZ01	MODULE	GEMFIRE CORP	NONE	NONE	1-May-98	1	G44196	169J265	2WMA521	12,991.02
9282704	WMZ01	MODULE	GEMFIRE CORPORAT	NONE	NONE	1-May-98	1	G44196	169J265	2WMA521	12,991.02
9282711	WMZ01	MODULE	GEMFIRE CORPORAT	NONE	NONE	1-May-98	1	G44196	169J265	2WMA521	12,991.03
9282728	WMZ01	MODULE	GEMFIRE CORPORATION		NONE	1-May-98	1	G44196	169J265	2WMA521	12,991.03
9282735	WMZ01	WAVEGUIDE	GEMFIRE CORPORAT	NONE	NONE	19-Aug-98	1	G44196	169J265	2WMA521	16,271.06
9282797	WMZ01	LASER	SPECTRA-PHYSICS L	3960-LIS	1738	12-Mar-98	1	G86152	169J265	2WMA521	50,606.88
9283466	WMZ01	WAVEGUIDE	GEMFIRE CORP	NONE	NONE	14-Aug-98	1	G44192	169J265	2WMA521	16,271.06
9119017	WMZ01	DRIVER	STANFORD	FABRICATI	207	11-Aug-97	1	G41051	169J265	2WMA521	5,434.16
											199,000.06

TOTAL 14