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.
Study of Photorefractive Effects in Periodically-Poled Lithium Niobate

Final Technical Report
for the period
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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 photorefractive 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.

![Crossed-beam setup](image)

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
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:

$$\Delta n_0 = \frac{32}{\pi^2} \frac{1}{(K_g w)^2} \Delta n_h$$

(1)

where $\Delta n_0$ is the index change at the center of the pump beam, $\Delta n_h$ 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 $\Lambda$ 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):

$$\Delta n_0 = a_0^2 \Delta n_h$$

(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 $L_{\text{eff}}$ in the medium.

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

$L_{\text{eff}}$ is given by:

$$L_{\text{eff}} = \frac{\pi}{\sqrt{2}} \frac{w}{n \sin \beta}$$

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

$$\Delta \varphi = 2\pi \Delta n L_{\text{eff}} / \lambda$$

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

$$\frac{\Delta I}{I} = \frac{1}{\sqrt{2}} \Delta \varphi$$

at a distance of $z = 2.72 \frac{w^2}{\lambda}$, where $\lambda$ 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 $t \to \infty$ 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 photolithographic 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](image)

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.](image)
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/cm² in the periodically-poled regions. This was reduced to 100W/cm² 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)

<table>
<thead>
<tr>
<th>period (µm)</th>
<th>nominal duty cycle - 40%</th>
<th>nominal duty cycle - 50%</th>
<th>nominal duty cycle - 70%</th>
<th>unpoled region</th>
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<td>0.006</td>
<td>0.25</td>
<td>0.15</td>
<td>0.05 (0.40)*</td>
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<td>0.006</td>
<td>0.055</td>
<td>0.25</td>
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<td>6.2</td>
<td>0.004</td>
<td>0.007</td>
<td>0.008</td>
<td>0.04 (0.32)*</td>
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<td></td>
<td>0.004</td>
<td>0.002</td>
<td>0.024</td>
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<tr>
<td>6.3</td>
<td>0.001</td>
<td>0.043</td>
<td>0.20</td>
<td>0.045 (0.36)*</td>
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<td>0.0035</td>
<td>0.024</td>
<td>0.030</td>
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<tr>
<td>6.4</td>
<td>0.004</td>
<td>0.055</td>
<td>0.10</td>
<td>0.12 (0.96)*</td>
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<td>0.005</td>
<td>0.024</td>
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<td>6.5</td>
<td>0.013</td>
<td>0.022</td>
<td>0.70</td>
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(*) data normalized to account for 8X reduction in pump power in unpoled regions.

The measurement of $\Delta I/I$ 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: $\Delta I/I$ in saturation

<table>
<thead>
<tr>
<th>period</th>
<th>nominal duty cycle - 40%</th>
<th>nominal duty cycle - 50%</th>
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<th>unpoled region</th>
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<tr>
<td>6.4</td>
<td>0.029 (1.1x10⁻³)*</td>
<td>0.155 (5.9x10⁻⁵)*</td>
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<td>6.5</td>
<td>0.087 (3.4x10⁻⁴)*</td>
<td>0.13 (5.0x10⁻⁵)*</td>
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</table>

(*) 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 $\Delta n / n$ is proportional to the slope $(1/l)\Delta n / dt$ for every measured value in Table 1, the steady-state index changes were calculated (Table 3).

Table 3: Steady-state index changes, $\Delta n_e \times 10^5$

<table>
<thead>
<tr>
<th>period (µm)</th>
<th>nominal duty cycle - 40%</th>
<th>nominal duty cycle - 50%</th>
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<th>unpoled region</th>
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<td>6.0</td>
<td>1.5 1.5</td>
<td>62 14 30</td>
<td>37 62</td>
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<td>6.2</td>
<td>1.0 1.0</td>
<td>1.7 0.50 0.25</td>
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<td>6.3</td>
<td>0.25 0.85</td>
<td>10 5.9 7.4</td>
<td>49 44</td>
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<tr>
<td>6.4</td>
<td>1.0 1.2</td>
<td>14 5.9 6.9</td>
<td>25 22</td>
<td>240</td>
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<tr>
<td>6.5</td>
<td>3.2 2.5</td>
<td>5.4 16 5.4</td>
<td>170 120</td>
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</table>

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:


## V. APPENDICES

### A. List of Equipment Purchased

**PROPERTY REPORT FOR CONTRACT F49620-97-1-0221**

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