THE PHOTOREFRACTIVE EFFECT IN LITHIUM NIOBATE

by

WILLIAM D. CORNISH

B.Sc. (Hon), Queen's University at Kingston, 1969
M.A.Sc., University of British Columbia, 1972

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We accept this thesis as conforming to the
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THE UNIVERSITY OF BRITISH COLUMBIA

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Department of Electrical Engineering

The University of British Columbia
2075 Wesbrook Place
Vancouver, Canada
V6T 1W5

Date 15 December, 1975
ABSTRACT

Exposure of the insulating ferroelectric crystal, lithium niobate, to light of the appropriate wavelength causes small changes in the refractive indices. This phenomenon which has recently been named the photorefractive effect allows phase holograms to be stored in the crystal. The work described in this thesis was undertaken to obtain an understanding of the mechanisms of the photorefractive effect in connection with possible engineering applications. The process is thought to involve the spatial redistribution of photo-excited electrons among traps. Space charge fields develop which modulate the refractive indices through the electro-optic effect.

Initially, the mechanisms proposed for charge transport were diffusion and drift in an internal field of pyroelectric origin. Using these mechanisms, Amodei had treated the initial development of phase holograms on the assumption that the electron transport length was short. A theoretical treatment without the restriction of short transport length is presented which shows that the efficiency of hologram writing increases for increased transport length up to a certain limit. In addition, it is shown that the resolution of the recording medium is not limited by increased transport length. More recently, Glass, von der Linde and Negran have proposed a new phenomenon, the bulk photovoltaic effect, as being responsible for charge transport. Photocurrent measurements are presented which provide further evidence for the existence of this effect.

The relative contributions of drift, diffusion and the bulk photovoltaic effect to the photorefractive process are investigated by applying a field during hologram formation. It is found that the effects
of positive and negative applied fields are not symmetric. The degree of asymmetry depends on what fraction of the crystal is illuminated. It is also found that both the voltages applied during previous exposures and voltages applied during the current exposure influence the diffraction efficiency. It is thought that these effects are caused by large scale space charge fields which are produced by exposure to light. The development of these space charge fields is discussed. It is concluded that the holograms were written by a combination of diffusion, drift in applied and space charge fields, and the bulk photovoltaic effect.

The importance of multiple internal reflections between the faces of the crystal had not previously been considered. It is shown that in measuring the photorefractive sensitivity by holographic, ellipsometric and adjustable-compensator techniques, neglecting multiple reflections may cause serious errors.

Two methods of probing large scale changes in the refractive indices are outlined. In the first method an automated ellipsometer was modified and programmed to measure the birefringence of the lithium niobate crystals and the change in birefringence due to illumination. This method has yielded information on the extent of optically-induced space charge fields, the uniformity of the crystal and the effects of heating crystals under different conditions. A second and more rapid method of inspecting large scale changes in the refractive indices is based on making the crystal act as a Fabry-Perot interferometer.

The sensitivity of photorefractive crystals to light has been associated with impurities and defects in the crystal lattice. Methods of modifying the valence state of iron impurities are important since
the photorefractive sensitivity is dependent on the amount of Fe$^{2+}$ present in the crystal and on the ratio of Fe$^{2+}$ to Fe$^{3+}$. One of the methods used to convert Fe$^{3+}$ to Fe$^{2+}$ is heating the crystal in lithium carbonate. It is shown that this treatment, in addition to reducing iron impurities, changes the birefringence of the crystal and reduces the rate at which space charge fields decay. It is suggested that the last effect is caused by the destruction of shallow traps.

A further charge transport mechanism has been suggested in which intervalence transfer of electrons between Fe$^{2+}$ and Fe$^{3+}$ impurity states occurs. It is argued that if, instead, electrons enter the conduction band, luminescence should be observable. It is shown that a photoluminescence band which is associated with iron impurities in the crystal may be observed in the region of 770 nm.
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To Diane
CHAPTER 1
INTRODUCTION

In certain insulating crystals, exposure to light of the appropriate wavelength induces small changes in the refractive indices*. This phenomenon has recently been named the photorefractive effect. Phase holograms may be stored in photorefractive crystals by exposure to two interfering coherent light beams. The photorefractive effect has been found in ferroelectric crystals such as lithium niobate (Ashkin et al. 1966), strontium barium niobate (SBN) (Thaxter 1969), barium titanate, lithium tantalate (Ashkin et al. 1966) and potassium niobate (Ostrowsky et al. 1970). In addition, the paraelectric crystal, potassium tantalate niobate (KTN) above its Curie point, exhibits the effect if an electric field is applied to the crystal (Chen 1967).

The work described here was undertaken to obtain an understanding of the mechanisms involved in the photorefractive effect in connection with possible engineering applications. Lithium niobate was chosen as the material for the study because it seemed the most promising material for applications and because high quality crystals are readily available. Although more was known about the photorefractive effect in lithium niobate than in other crystals, the basic mechanisms were not understood, the specification of the material for engineering applications was incomplete and it was not clear how to optimize the properties of the crystal.

* More precisely, the light induces a change in the optical indicatrix.
When lithium niobate first became available in single crystals of optical quality, its high electro-optical coefficients and other properties made it attractive for use in electro-optic modulators. It was found, however, that illumination caused optical inhomogeneities to form in the crystal and these degraded the performance of the modulator. The crystals were most sensitive to the blue and green wavelengths of light. The objective of the first investigations was to find ways of eliminating the photorefractive effect. Control of impurities in the crystal, especially iron, was found to be important in this regard.

Chen, LaMacchia and Fraser (1968) were the first to point out that the photorefractive effect can be used to store thick phase holograms in lithium niobate. In contrast to photographic methods, no development or bleaching processes are required. The holograms can be optically or thermally erased and new holograms written. Diffraction efficiencies approaching 100% are theoretically possible. Experimentally, diffraction efficiencies of up to 60% have been reported (Amodei et al. 1972).

Chen et al.'s discovery created interest in the potential application of lithium niobate as a holographic storage medium in optical computer memories. This application is being investigated by several companies. It was shown by van Heerden (1963) that theoretically the ultimate storage capacity of a volume hologram is \( \frac{V}{\lambda^3} \) bits where \( V \) is the volume and \( \lambda \) is the wavelength of light. This means that theoretically more than \( 10^{12} \) bits can be stored in a 1 cm\(^3\) crystal.

One method of optically storing data is on a page by page basis. Each page containing \( N \) bits would be written or read as a
hologram. A large number of holograms would be stored either in an x-y array or in superposition. For a writing time of $\tau$ sec, the writing rate would be $N/\tau$ bits/sec. For instance, if hologram storage of a page containing $10^4$ bits required 1 ms, the writing rate would be $10^7$ bits/sec.

Another method was suggested by Carlsen (1974) which would not require the page of data to be assembled before storage. Presently, only prototype page composers have been built. The data could be written one bit at a time in an x-y array with random access to any bit on a page. Data would be read out in parallel, one page at a time. Experimentally, Carlsen has stored pages of 16,000 bits in iron-doped lithium niobate.

A number of review articles are available which discuss the advantages and limitations of optical memories (Rajchman 1970, King 1972, Anderson 1972, Hill 1972, Kiemle 1974, Chen and Zook 1975). (See Appendix E).

It is believed that the mechanism of the photorefractive effect may be broadly described as follows. Exposure of a photorefractive crystal to light of the appropriate wavelength causes photo-excitation of electrons from traps. A spatial redistribution of the electrons among traps sets up space charge fields which affect the refractive indices via the electro-optic effect. The space charge fields may be altered by further exposure to light. The optical inhomogeneities may be removed by heating the crystal to approximately 200°C. Electrons are thermally excited and uniformly distributed so that on cooling, the fluctuations in the refractive indices are removed. The mechanisms which redistribute the electrons are of
primary interest in current research.

Rapid development of the subject occurred during the course of this study. At its beginning, it was recognized that both drift and diffusion were possible transport mechanisms (Amodei 1971a, 1971b) but their relative importance was not established. A theoretical treatment of the development of refractive index gratings through drift or diffusion was made without the restriction of a short migration length as assumed by Amodei (1971a). This is discussed in Chapter 2. It was shown that the efficiency of hologram writing increases for increased migration length up to a certain limit. Also, increased migration length would not limit the resolution of the recording medium.

Glass, von der Linde and Negran (1974b, 1975a) have proposed that the photorefractive effect involves an entirely new transport mechanism which they have labelled the "bulk photovoltaic effect". This mechanism is thought to be responsible for photocurrents which were previously attributed to internal fields of pyroelectric origin. The bulk photovoltaic effect is outlined in Chapter 2 and in later chapters (7,8,9) experimental results which bear upon the effect are discussed.

In measuring the diffraction efficiency of phase holograms stored in insulating crystals, the effect of multiple internal reflections between the crystal faces had been neglected. Measurements of the build-up of the diffraction efficiency with time have been used to test various physical models for the photorefractive effect. Usually a prototype hologram has been written by the interference of two plane waves. It is then necessary to relate the observable diffraction efficiency to the predicted refractive index modulation. The effects of multiple reflections are discussed in Chapter 5. It is shown that neglecting multiple reflections can cause serious errors. Small changes in temperature (approximately 1 °C) such as those that are
easily produced by exposure to medium intensity laser beams produce sufficient change in crystal thickness to produce significant effects.

In Chapter 7, the effects of applying an external field to the crystal are investigated. This is a useful method of probing the transport mechanism and may also be a useful means of controlling hologram formation. The published data obtained from using this technique were in apparent contradiction. It is shown that the results of such an experiment depend on the field applied during previous exposure to light as well as on the field applied during the current experiment. It is also shown that the portion of the crystal illuminated will determine the magnitude of the space charge field that may develop in the crystal. Different illumination geometries give different results. Since the applied field changes the optical thickness of the crystal, the effect of multiple reflections may also affect the results.

Observations of optically induced changes in the birefringence due to single beams can produce useful information. The compensator method of Chen (1969) is slow and does not allow for absorption. It is shown (Chapter 3) that ellipsometry has certain advantages and its usefulness has been considerably increased through automatic computer control. It is shown that, as in hologram studies, the effects of internal multiple reflections must be taken into account in the ellipsometric and adjustable compensator methods. In addition to using the ellipsometer, it is shown that large scale changes in the refractive index can be rapidly inspected if the crystal is made to act as a Fabry-Perot interferometer (Chapter 4).

The sensitivity of photorefractive crystals to light has been associated with impurities in the crystals and defects related to the
non-stoichiometry of the crystal (Peterson et al. 1972). It is of primary importance to determine the nature and properties of the defect sites from which electrons may be photo-excited and trapped. The most efficient method found to increase the photorefractive sensitivity has been the addition of iron impurities (Phillips et al. 1972, Peterson et al. 1973).

Phillips and Staebler (1974) have shown that heating lithium niobate crystals while packed in Li$_2$CO$_3$ reduced iron impurities from Fe$^{3+}$ to Fe$^{2+}$. They have used this method to control photorefractive sensitivity (Staebler and Phillips 1974a). Crystals treated by this method have been studied with the ellipsometer (Chapter 3) and by holographic methods (Chapter 7). It is shown that in addition to reducing the iron impurities, the treatment changes the birefringence of the crystal and decreases the rate at which the optically-induced space charge fields decay. To explain these results it is proposed that the treatment destroys shallow traps.

To determine whether electrons enter the conduction band or whether charge transport is the result of intervalence transfer, a search for luminescence was made. A luminescence band at 770 nm was observed (Chapter 8) which was associated with the iron impurities in the crystal. The luminescence was stronger in crystals after heat treatment in Li$_2$CO$_3$. 
CHAPTER 2
PHYSICAL MODELS FOR THE PHOTOREFRACTIVE EFFECT

2.1 Introduction

As was mentioned in the previous chapter, a number of models have been proposed to explain the photorefractive effect. The development of these models is now outlined to show how they affect the present understanding of the process.

2.2 The Electro-optic Nature of the Photorefractive Effect

Chen, LaMacchia and Fraser (1968) found that the reconstruction of holograms stored in lithium niobate was only 1/10 as efficient for ordinary ray illumination as for extraordinary ray illumination. To explain this observation, they suggested that the photorefractive process responsible for hologram storage involved the electro-optic effect in the crystal.* The diffraction efficiency $\eta$ is proportional to $\sin^2(a\Delta n)$, where $a$ is a constant and $\Delta n$ is the amplitude of the refractive index modulation (Kogelnik 1969). This implies that for Chen et al.'s observation $(\Delta n_e)/(\Delta n_o) \approx 0.3$. This is consistent with the electro-optic coefficient $r_{13}$ which affects the ordinary index being 1/3 that which affects the extraordinary index $r_{33}$.

In contrast with this, Gaylord et al. (1972) observed no dependence of the diffracted power on the polarization of the readout beam. They have since found (private communication) that this observation was peculiar to one particular crystal and all other measurements

* Appendix 2 contains an outline of the electro-optic effect in lithium niobate.
they have made are consistent with Chen et al.'s observations. They speculated that the holograms they saw were not caused by the mechanisms usually associated with the photorefractive effect.

To justify using the relative magnitudes of $r_{13}$ and $r_{33}$ to account for the polarization dependence of reading holograms, Chen et al. proposed that the refractive indices were modulated by a space charge field directed along the $x_3$ or c-axis of the crystal. The next section details further experiments conducted by Chen to verify this.

2.3 The Internal Field Theory

Using an adjustable compensator method (Sec. 3.1), Chen (1969) observed changes in birefringence induced with a single laser beam. Fig. 2.1(a) shows the optically induced change in birefringence along lines parallel and perpendicular to the c-axis. The birefringence reverses sign along the c-axis but not along the b-axis. To explain this observation, Chen proposed a model in which an electron could be excited from one trap and then captured in another. He assumed that there was an internal electric field $E_0$ directed from the positive end of spontaneous polarization of the crystal to the negative end, that is, antiparallel to the spontaneous polarization vector $P_s$. The direction of the field was determined by applying an external field to the crystal. One direction of the field retarded the change in the birefringence while the other direction enhanced the change. This field would cause photo-excited electrons to drift along the c-axis toward the beam periphery. Chen appears to have envisioned that after being retrapped and reexcited many times, these electrons would drift out of the illuminated area and would remain trapped at levels too deep to be
Fig. 2.1 (a) The solid line (---) shows the change in birefringence along the c-axis and the dashed line (-----) the change along the b-axis due to a beam of circular symmetry ($\lambda = 488$ nm). (b) Chen's postulated space charge field distribution which causes the observed change in $\Delta(n_e - n_o)$. 
reexcited by thermal processes. The space-charge field, $E_{sc}$, created by the trapped electrons and positively ionized centers from which the electrons originated caused the observed refractive index variation via the electro-optic effect (Fig. 2.1(b)). Since lithium niobate exhibits a linear electro-optic effect, the variation $\Delta(n_e - n_o)$ is linearly related to the spatial variation of the electric field. The space charge field required for the magnitude of the observed effect was $6.7 \times 10^4$ V/cm.

To verify that there was an internal field residing in the crystal, Chen looked for and found a short circuit photocurrent. The direction of the photocurrent was consistent with a field opposite to $P_s$. It was suggested that the field might be of pyroelectric origin since a portion of the light used to form the hologram would cause non-uniform heating of the crystal. However, Chen pointed out that $(dP/dT < 0)$ and therefore the field would be in the wrong direction for his observations. Although it seemed evident that an internal field existed, Chen did not account for its origins.

### 2.4 Johnston's Polarization Model

To remove the need to assume a built-in field of unknown origin, Johnston (1970) proposed an alternative model in which photoinduced variations in the macroscopic polarization caused the photorefractive effect. Illumination of the crystal would excite electrons in the conduction band resulting in a change in the density of filled traps in the region of illumination. This in turn would cause a local change in the polarization. The divergence of the polarization produces a field which skews the diffusional process of electrons in
the conduction band. After the light is turned off, there remains a change in the macroscopic polarization which induces a change in the refractive indices of the crystal.

Using this model, Johnston was able to account qualitatively for the spatially dependent features of Chen's observations (Fig. 2.1). However, Amodei (1971a) and Amodei and Staebler (1972b) have shown that there are a number of difficulties with this mechanism. They concluded that a very large number of electrons would be required to enter the conduction band to account for the generation of the fields necessary to skew the electronic motion. The same magnitude of induced index change could result from space charge fields created through simple diffusion and retrapping processes. The number of electrons involved would be less by a factor of $10^3$ than would be required in Johnston's model. In addition, they observed that Johnston's resistivity measurement ($10^{13}$ ohms) which led him to conclude that any internal fields would relax in a short time, was abnormally low. Typical values would allow fields to remain for many weeks.

Amodei and Staebler (1972b) suggested that the built-in field which Chen used to explain his results was of pyroelectric origin and developed when the crystal was cooled from a high temperature. The development of such a field may be explained in the following way.

A rectangular $3m$ ferroelectric crystal with no free charges and no net space charges would have a field corresponding to a polarization charge $P_{rem}$ per unit area on faces normal to the c-axis. This would, in fact, be above normal dielectric breakdown values. In practice, the crystal would have been cooled from some high temperature at which appreciable conductivity existed, sufficient to cancel the field. Excess
charges would accumulate close to each c-face. As cooling progresses, the conductivity will freeze out while the remanent polarization \( P_{\text{rem}} \) continues to change. Finally, an uncompensated component of \( P_{\text{rem}} \) will exist giving a built-in field of magnitude

\[
\frac{1}{\varepsilon} \int_{T_0}^{T_1} \frac{\partial P_{\text{rem}}}{\partial T} \,dT
\]

where \( T_1 \) and \( T_0 \) are the temperatures at which the conductivity disappears and the temperature of the experiment respectively, and \( \varepsilon \) is the permittivity.

2.5 The Formation of Holograms by Drift or Diffusion

2.5.1 Introduction

To probe the mechanism of hologram storage in lithium niobate, it is convenient to analyze the formation of a prototype hologram formed by causing two coherent plane waves to interfere in the volume of the crystal. The resulting interference pattern is sinusoidal and the intensity is of the form

\[
I = I_0 (1 + m \cos kx)
\]

(2.1)

where \( k \) is the spatial frequency of the pattern, and \( m \) is the modulation ratio* and \( x \) is in the plane of the two beams and perpendicular to their bisector. For two beams intersecting at an angle \( 2\theta \) as shown in Fig.2.2

\[
k = \frac{2\pi}{\lambda} \quad \text{where} \quad \lambda = \frac{\lambda}{2 \sin \theta} \quad \text{and} \quad \lambda \text{ is the wavelength of the light.}
\]

Amodei (1971a) has used this configuration to analyze the formation of holograms. He assumed that the drift or diffusion length of photo-excited electrons was very much smaller than the grating period \( \lambda \).

* A definition of the modulation ratio is given in Chapter 5.
Fig. 2.2 Configuration for recording holograms. The two plane waves R and S interfere to produce a sinusoidal light intensity pattern with a period \( \lambda \). The c+ end of the crystal (of thickness d) is shown in the positive x direction.
Staebler and Amodei (1972b) have shown that depending on the mechanism of charge transport, the periodic index modulation may be shifted in phase with respect to the intensity modulation that created it. Observations on the transfer of energy between the two writing beams, and the assumption that free electrons moved a very short distance before being retrapped (Amodei 1971b) led them to conclude that both diffusion and drift were responsible for the grating formation. In Sec. 2.5.4 it is shown that the interpretation of their results depends on how far electrons move.

2.5.2 Analysis for Short Drift or Diffusion Length

Chen et al. (1968) found that they could store holograms with a resolution of greater than 1600 lines/mm. This led them to assume that the displacement of electrons due to the illumination must be a fraction of a micron to be able to record the variation in intensity. Because of this, Amodei assumed that in developing a theory, it would be reasonable to restrict the drift or diffusion length to a fraction of the grating wavelength.

Amodei assumed that the rate of promotion of electrons into the conduction band \( g(x) \) to be proportional to the intensity of light, at least in the initial stages of hologram formation when the traps may be taken as uniformly filled. The assumption of short drift or diffusion length implies that the free-carrier concentration is given by

\[
n(x) = g_0 \tau (1 + m \cos kx)
\]

where \( \tau \) is the lifetime of carriers and \( g_0 \) is proportional to \( I_0 \) (in Eq. 2.1). The spatial distribution of the current was taken as the sum of the drift and diffusion components,
\[ J(x) = \mu e E(x) + eD \frac{dn(x)}{dx} \quad (2.3) \]

where \( e \) is the electronic charge, \( \mu \) is the mobility for electrons, \( E(x) \) is the total electronic field and \( D \) is the diffusion constant for electrons. The rate at which space charge density, \( \rho \), accumulates at any point is given by the continuity equation

\[ \frac{d\rho}{dt} = -\nabla \cdot J. \quad (2.4) \]

Combining this with Eq. 2.3 the build-up of the space charge density can be expressed as

\[ \rho(t) = \int_0^t \int \left( \mu e E + eD \frac{dn}{dx} \right) \, dt \quad (2.5) \]

The space charge field supported by the space charge density is

\[ E_{sc}(x) = \int \frac{\rho}{\varepsilon} \, dx \quad (2.6) \]

where \( \varepsilon \) is the dielectric constant of the material.

In Eq. 2.4, the space charge density \( \rho \) which Amodei has used to calculate the space charge field, includes not only the trapped charge density, but also the free charge density in the conduction band. These equations, then, describe the situation during illumination. After illumination ceases the free electron density decays.

Amodei considered (i) transport due to drift only, \( J = ne\mu E \), where \( E = -E_o \) (the space charge field \( E_{sc} \) is neglected in the transport equation, and the built-in field is taken negative to cause electrons to drift in the positive \( x \) direction); and (ii) transport due to diffusion only \( J = eD \frac{dn}{dx} \). Solution of Eqs. 2.4 to 2.6 yield for drift only

\[ E_{sc} = \frac{(e\mu E_0 t_0 m) \cos kx}{\varepsilon} \quad (2.7) \]
and for diffusion only,

\[ E_{sc} = \frac{(eDtg_{0} mk) \sin kx}{\varepsilon} \]  

Thus, with the above assumption of short drift or diffusion length, a response is obtained with a difference of \( \pi/2 \) in phase shift according to whether drift or diffusion is operative.

It is interesting to note (Young et al. (1974)) that the shorter the diffusion or drift lengths are assumed to be, the slower the production of the index grating since the electrons are assumed to return more nearly to their original positions.

2.5.3 The Effects of Beam Coupling

Staebler and Amodei (1972b) have considered the implications of beam coupling during reading and writing holograms. They have shown that coupled wave analysis allows the determination of whether the periodic index gratings are shifted with respect to the periodic intensity patterns which produce them. In the case they considered, two coherent beams \( R \) and \( S \) are symmetrically incident at an angle \( \theta \) relative to the \( z \) axis as shown in Fig. 2.2. Both waves are polarized perpendicular to the plane of incidence and are incident on a region with periodic variations of refractive index,

\[ \Delta n = n_{1} \cos kx \]  

that extends from \( z = 0 \) to \( z = d \). The two waves \( R \) and \( S \) can be written in the form

\[ R = r(z) \exp(-i(2\pi \cos \theta z + kx)) \]
\[ S = s(z) \exp\left(-i \frac{2\pi \cos \theta z}{\lambda} - kx\right) \]  

Kogelnik (1969) (see Appendix C) using coupled wave theory has shown that for a nonabsorbing phase grating and perfect Bragg conditions,

\[ \frac{dr(z)}{dz} = -i\kappa s(z) \]  

\[ \frac{ds(z)}{dz} = -i\kappa r(z) \]

where \( \kappa = \frac{\pi n_1}{\lambda \cos \theta} \).

The coupled wave equations have the general solution

\[ r(z) = a \exp \left(i\kappa z\right) + b \exp \left(-i\kappa z\right) \] \[ s(z) = -a \exp \left(-i\kappa z\right) + b \exp \left(i\kappa z\right) \]

If we let the boundary conditions be \( r(0) = 1 \) and \( s(0) = A \exp \left(-i\phi\right) \) then the coefficients \( a \) and \( b \) can be determined and the beam amplitudes become

\[ r(z) = \cos \left(\kappa z\right) - iA \exp \left(-i\phi\right) \sin \left(\kappa z\right) \] \[ s(z) = -i \sin \left(\kappa z\right) + A \exp \left(-i\phi\right) \cos \left(\kappa z\right) \]

The intensities of the two beams are given by

\[ I_R = |r|^2 = \cos^2 \left(\kappa z\right) + A^2 \sin^2 \left(\kappa z\right) - A \sin \left(2\kappa z\right) \sin \phi \] \[ I_S = |s|^2 = \sin^2 \left(\kappa z\right) + A^2 \cos^2 \left(\kappa z\right) + A \sin \left(2\kappa z\right) \sin \phi \]

If the beams have equal incident amplitudes, then \( A = 1 \) and Eq. 2.13 gives

\[ I_R = 1 - \sin \left(2\kappa z\right) \sin \phi \] \[ I_S = 1 + \sin \left(2\kappa z\right) \sin \phi \]

Staebler and Amodei (1972b) have shown that these equations also describe the situation where the two waves have a fixed phase relationship, but the phase grating is moveable along the x axis with \( \Delta n = n_1 \cos \left(kx + \phi\right) \). Eq. 2.14 now indicates that the energy transfer
between beams depends on the position of the grating with respect to the interference pattern. The phase factor \( \phi \) in Eq. 2.14 represents the phase shift between the light intensity pattern and the index modulation it produces.

2.5.3.1 Coupling During Writing

Using the experimental arrangement shown in Fig. 2.3, Staebler and Amodei (1972b) found that during hologram formation there was energy transferred between the beams. From Amodei's (1971) analysis of the transport process during recording (sec. 2.5.2), holograms formed by diffusion produce a phase shift \( \phi = \pi /2 \) and holograms formed by drift produce a phase shift \( \phi = 0 \). Eq. 2.14 indicates that only the values of \( \phi \) other than zero or \( \pi \) cause energy transfer between the beams. This was taken as evidence for holograms stored by diffusion.
2.5.3.2 Interactions During Readout

When a previously recorded hologram is read out, it is possible for a new hologram to be written by the interference of the reading beam and the diffracted beam. During readout, the S beam is blocked so that the boundary conditions are \( s(0) = 0 \), and \( r(0) = 1 \). From Eq. 2.12, the amplitude of the two beams within the diffraction grating \( n = n_1 \cos(kx) \) are given by

\[
\begin{align*}
r(z) &= \cos(\kappa z) \\
s(z) &= -i \sin(\kappa z)
\end{align*}
\]

(2.15)

The interference pattern produced by these two beams is found by substituting Eq. 2.15 into Eq. 2.10 with the result

\[
I_{\text{total}} = |r + s|^2 = 1 + \sin(2\kappa z) \sin(kx)
\]

(2.16)

If the mechanism of hologram formation produces an index modulation proportional to the intensity then the new grating \( \Delta n_2 \) will be

\[
\Delta n_2 = n_2 \sin(kx)
\]

(2.17)

The total phase modulation will be

\[
\Delta n_t = n_1 \cos(kx) + n_2 \sin(kx)
\]

(2.18)

Staebler and Amodei (1972b) argued that the effect of \( n_2 \) was to bend the phase grating. This effect on the total diffraction efficiency was not pursued except to point out that the effect should be the same for readout with the R beam or the S beam.

In the case where hologram formation produces an index modulation shifted by \( \pi/2 \), the new grating \( \Delta n_3 \) will be

\[
\Delta n_3 = n_3 \cos(kx)
\]

(2.19)

The total phase modulation will be
\[ \Delta n_t = (n_1 + n_3) \cos (kx) \]  

(2.20)

The effect of \( n_3 \) is to increase or decrease the effectiveness of the grating. Although the value of \( n_3 \) will be larger toward the back of the grating, the diffraction efficiency depends only on the amplitude of the grating (Kermish 1969). An example of this is shown in Fig. 2.4. Curve 3 illustrates a phase grating that resulted from diffusion of electrons. The space charge and the space charge field producing this grating are shown in curves 1 and 2 respectively. It is assumed that a positive field decreases the index when the c-axis is oriented as shown. When the R beam is used for readout, it interferes with the diffracted beam to form a light-intensity pattern shown by the fourth curve. The intensity maximum is on the +x side of the peaks in curve 3 because the R beam is propagating in the +x direction. The fifth and sixth curves show the space charge and the space charge field resulting from diffusion of electrons due to the intensity pattern of curve 4. The induced index change \( \Delta n_3 \) will either enhance or degrade the original index pattern \( \Delta n_1 \) depending on the direction of the +c-axis. If the crystal is oriented in the same way as it was for the production of \( \Delta n_1 \), then enhancement occurs. If the crystal is reversed so that the +c end of the crystal is facing in the -x direction, the total diffraction efficiency will be degraded. The same result is obtained if the crystal is not reversed but the S beam is used for readout. This reverses the phase of the light modulation, and the space charge that accumulates opposes the space charge already present.

Staebler and Amodei's coupled-wave analysis has shown that interaction between the two beams used in holography depends on the sine of the phase angle \( \phi \) between the intensity modulation and the index
Fig. 2.4 (1): sinusoidal space charge created by a sinusoidal light pattern. (2): the space charge field created by (1). (3): variation in the refractive index caused by (2) through the electro-optic effect. (4): a second light intensity pattern which creates (5), a space charge and (6), a space charge field. The space charge field either enhances or degrades the grating $\Delta n_1$ depending on the direction of the +c end of the crystal.
modulation. Using Amodei's treatment of the formation of phase holograms, they restrict the value of $\phi$ to $\pi/2$ when diffusion is responsible for charge transport and to zero or $\pi$ when drift is responsible for charge transport. It is not obvious why $\phi$ should be such a discontinuous function. Physically it would be more reasonable for $\phi$ to vary smoothly. In the next section it is shown that if the restriction of short drift or diffusion length is removed, then drift will allow any value of $\phi$ depending on the drift length where as the value of $\phi$ for diffusion will be $\pm \pi/2$ independent of the diffusion length.

2.5.4 Analysis with Arbitrary Drift or Diffusion Length

The need for the assumption that the drift or diffusion length be short may be removed by using the continuity equation (Young, Wong, Thewalt and Cornish 1974)

$$\frac{\partial n}{\partial t} = g - n + \frac{1}{\tau} \frac{\partial J}{\partial x}$$  \hspace{1cm} (2.21)

In the initial stages of hologram formation, the rate of change of the concentration of free electrons in the conduction band is zero at constant light intensity so that

$$0 = g - n + \frac{1}{\tau} \frac{\partial J}{\partial x}$$  \hspace{1cm} (2.22)

Amodei's (1971a) assumption that $n = g^+$ corresponds to dropping the term $(1/e) \partial J/\partial x$ in Eq. 2.22. This term is the negative of $g - n/\tau$, which gives the rate of trapped space-charge build-up causing the effects of interest. For the drift only case, $J(x) = ne\nu E$ where $E = -E_0$. Writing $g = g_0(1 + m \cos (kx))$, Eq. 2.22 becomes

$$\frac{dn}{dx} + \frac{n}{L} = \frac{g_0}{\nu E_0} (1 + m \cos kx)$$  \hspace{1cm} (2.23)
where $L = \mu E_0^\tau$ and $g_o$ is proportional to $I_0$ (Eq. 2.1). This may be solved to yield

$$n(x) = \tau g_o u(x) + (n(0) - \frac{\tau g_o m}{1 + L^2 k^2}) \exp(-x/L)$$

$$+ \frac{\tau g_o m}{1 + L^2 k^2} (\cos kx - kL \sin kx) \quad (2.24)$$

where $u(x)$ is a unit step function and $n(0)$ is the initial value of the free electron concentration. Then with $\partial \rho / \partial t = -\partial J / \partial x$ and $\partial E_{sc} / \partial x = \rho / \epsilon$, the space charge field may be written

$$E_{sc} = \frac{\epsilon}{e \epsilon \tau} \int (g \tau - n) \, dx \quad (2.25)$$

Substituting in Eq. 2.25, and excluding terms due to the termination of the periodic light pattern, the form of the space charge field is

$$E_{sc} = \frac{\epsilon g_o m}{\tau k} \left( \frac{k^2 L^2}{1 + k^2 L^2} \sin kx + \frac{kL}{1 + k^2 L^2} \cos kx \right) \quad (2.26)$$

This equation can also be derived by taking the convolution of the light pattern with the impulse response which, for this case, is proportional to $\exp(-x/L)$. (In the diffusion case, we have two exponentials back-to-back.) Physically we have a positive space charge wave due to the removal of electrons from traps plus a negative space charge wave due to their retrapping.

For the diffusion-only case, $J = eDdn/dx$. Writing $L' = (D\tau)^{1/2}$, the solution for the space charge field due to diffusion is

$$E_{sc} = \frac{e g_o m k L'}{\epsilon (1 + k^2 L'^2)} \sin kx \quad (2.27)$$
2.5.5 Discussion

For the case of space charge fields developed by drift, the phase shift between the periodic light intensity pattern and the periodic space charge field depends on the drift length. In Eq. 2.26, for \( L_k \ll 1 \), Amodei's expression is obtained (with \( E_{sc} \propto \cos kx \)). For \( L_k \gg 1 \) a much larger space charge field is obtained given by

\[
E_{sc} = (\text{teg} m/\varepsilon k) \sin kx.
\]

The response for this case is due to the positive space charge wave only, the negative charge due to retrapped electrons being uniform. It is also independent of \( E_0 \) and \( \tau \). The phase shift between the light intensity pattern and the index grating produced depends on the magnitude of the drift length \( L \) and varies from zero (for \( L_k < 1 \)) to \( \pi/2 \) (for \( L_k \gg 1 \)).

In the case of diffusion-formed space charge fields, for large enough \( L'k \), the same limiting case is obtained as for drift only with \( L_k \gg 1 \). However, whatever the magnitude of \( L'k \) the phase shift remains the same because the free electrons have equal probability of moving in either direction.

The magnitudes of \( L \) (for the drift case) to which Staebler and Amodei's results on coupled wave analysis apply are crucial. A phase shift halfway between the two limiting cases is achieved when \( L_k = 1 \). The drift length is then given by

\[
L = E_0 \mu \tau = 1/2 \pi = \lambda / 4 \pi \sin \theta.
\]

For \( \theta = 15^\circ \) and \( \lambda = 500\text{nm} \) a drift length of 153.7 nm is calculated. (For \( \theta = 45^\circ, L = 56.3 \text{ nm} \). The actual value of \( L \) will depend on the distance between empty traps, and their cross section. Taking \( E_0 = 10^4 \text{ V/cm} \) (which is smaller than Chen's (1969) estimate of the pyroelectric field in his crystals) and \( \mu = 15 \text{ cm}^2/\text{Vsec} \) (extrapolated from 1000°K (Jorgensen et al. 1969)), a value of \( \tau = 10^{-10} \text{ sec} \) is obtained. The
pyroelectric field would be expected to be rather an uncontrolled quantity which depends on the history, in particular the thermal history, of the crystal so that the value of L would depend on the particular specimen used.

It has been suggested by Staebler and Amodei (1972b) that the physical processes involved in the photorefractive effect due to single light beam may not necessarily be the same as operate in hologram formation. More specifically, the higher the spatial frequencies involved in the light intensity pattern, the more diffusion will tend to become important. The question may be tested by considering a hologram formed by two plane waves. Taking, for simplicity, both diffusion length L' and drift length L to be short compared to the reciprocal of the spatial frequency, the space charge field due to drift would be small compared to that due to diffusion if $E_o << Dk$. Using the Einstein relation between $\mu$ and D ($D = K'T\mu/e$) for a wavelength $\lambda = 500\text{nm}$, temperature $T = 300^\circ\text{K}$ and $\theta = 6.75^\circ$ (calculated from an angle of incidence of $15^\circ$), $E_o << K'Tk/e = 765 \text{ V/cm}$, where K' is the Boltzmann constant. For diffusion to dominate, the total field in the crystal would have to be only a few 10 V/cm. If the drift length were not negligible compared to 1/k, the field would have to be even smaller. Thus a rather complete suppression of the total field would be required.

In conclusion, an increase in drift or diffusion would cause an increase in writing efficiency up to a certain limit. Also, in deciding whether drift or diffusion is operative in a particular experiment, evidence based on detecting a spatial shift between the refractive index grating and the sinusoidal light intensity which produced it must be interpreted bearing in mind the following considerations. A grating produced by diffusion will be shifted by $\pm \pi/2$ in the reference frame.
defined by the light beams, the sign depending on the direction of the c(+) axis with respect to the light beams. For a grating produced by drift, the shift depends on the drift length \(L\). For \(L \gg l/k\), the shift is the same as for diffusion. For \(L \ll l/k\), there will be either 0 or \(\pi\) shift depending on the direction of the field causing drift, with respect to the c(+) direction and the sign of the appropriate electro-optic coefficient. If the field is a built-in field of pyroelectric origin, then changing the direction of the c(+) axis also reverses the field and so has no effect on the direction of electron motion with respect to the crystal axes. Intermediate values of \(L\) give intermediate shifts. In general, other evidence will be required to decide the point, for example, tests of the effect of an applied field such as that reported by Staebler and Amodei (1972b).

Recently, Vahey (1975) has presented a nonlinear coupled-wave theory of hologram storage in LiNbO\(_3\). Closed-form solutions are found to describe the energy exchange between beams during recording. The time development of the diffraction efficiency is also described. Using the concept of Sec. 2.5.4 (Young et al. 1974) which involves the dependency of the phase shift between the phase grating and the light interference pattern on the drift length of electrons, Vahey's equations provide a good description of Staebler et al.'s (1972b) coupled-wave experiments.

2.6 The Bulk Photovoltaic Effect

Glass, von der Linde and Negran (1974b, 1975a) have proposed that the photorefractive effect is caused by an entirely new transport mechanism which they have labelled the "bulk photovoltaic effect".
Chynoweth (1956) and Chen (1969) observed that photocurrents would flow in BaTiO$_3$ and LiNbO$_3$ respectively, in the absence of applied fields. In both cases the effect was accounted for by internal fields due to space charge effects. Glass et al. (1974b) stated that after 20 hours of continuous illumination, the photocurrents which they measured remained constant. The photoconductivity would relax any internal fields and a decay of the photocurrent would be noticeable. Experimental observations of the photocurrent are presented in Chapter 7. Glass et al. have used the bulk photovoltaic effect rather than internal fields to account for the photocurrent. Although the physics of this new phenomenon perhaps have not been fully established, it appears that Glass et al. are justified in proposing that it exists.

Glass, von der Linde and Negran have postulated that electrons contributing to the photocurrent and the photorefractive process reside in asymmetric potential wells (Fe$^{2+}$ ions). Upon excitation, there is a greater probability that they will move in one direction than another. They account for this phenomenon by assuming that the Nb-Fe$^{2+}$ distances in the ±c-direction are different. The asymmetry at all the defects has the same effect and there is a net electronic current $J_{el}$. Following the excitation of the electron, the ionized impurity is displaced along the polar axis of the crystal due to Franck-Condon relaxation. This gives rise to a displacement current $J_{e2}$. After a certain time the electronic momentum will become random and no longer contribute to the current until recombination. Glass et al. also suggest that the recombination process may be asymmetric. The probabilities of recombination from electrons approaching a trap from the ±c-direction are different and a net recombination current $J_r$ arises. After recombination the impurity
moves back to its original position but this process contributes no current since the impurity and captured electron move together. The total steady-state current $J$ is given by

$$J = J_{e1} + J_{e2} - J_r = \kappa\alpha I$$

(2.28)

where $\alpha$ is the absorption and $I$ the intensity of light. The term $\kappa$ is dependent on the nature of the absorption centre (the directional probabilities and mean free paths of electronic motion on excitation and recombination) and the photon energy. Even if the net electronic current created by excitation and recombination is zero, there will still be a net current due to the ionic displacement $J_{e2}$. On open circuit, the current $J$ will charge the crystal

$$J = \kappa\alpha I + e\mu E$$

(2.29)

so that a field $E$ will be created which will saturate at $E_{\text{sat}} = \kappa\alpha I/e\mu$.

### 2.7 Transient Photorefractive Effects

In addition to the previous models which attempt to explain optical inhomogeneities which persist long after the illumination is removed, Glass et al. (1975a, 1975b) have studied a photorefractive process that persists for only a very short time ($\sim 10\mu$sec). They suggest that the process is caused by a variation in the polarization. Thermal excitation of the crystal results in a pyroelectric polarization due to thermal expansion. Optical excitation produces a polarization change due to a change of the dipole moment of the excited defect. These processes produce a displacement current $J = dD_e/dt$, where $D_e$ is the electronic displacement. The lifetime of the effect is determined by the relaxation time of the excited state. These transient effects have been observed in LiNbO$_3$ doped with Cr$^{3+}$ ions (Glass et al. 1975a, 1975b) but not
in "undoped" or iron-doped lithium niobate.

2.8 Defect Sites

The photorefractive effect is most efficient when light of the wavelengths 400 to 500nm is used (Serreze and Goldner 1973). Excitation is thought to occur from traps within the 3.72 ev band gap (Clark et al. 1973). As was mentioned in Chapter 1, both impurities and defects related to the non-stoichiometry of the crystal act as defect sites. Phillips et al. (1972) have shown that the gamma irradiation (of un-stated energy) of undoped LiNbO₃ increases the photorefractive sensitivity by increasing the concentration of lattice defects which act as electron traps. In addition, impurity doping with elements such as iron, manganese, copper, rhodium and chromium (Phillips et al. 1972, Peterson et al. 1971, 1973; Mikami et al. 1973, Glass et al. 1974) improve the sensitivity of the crystal.

As was pointed out in Chapter 1, iron has so far proven to be the best dopant. Studies of nominally pure lithium niobate have revealed the presence of iron contamination of 10 ppm to 100 ppm in all samples tested (Peterson et al. 1971, 1973; Nash 1973). These impurities are thought to be chiefly responsible for the photorefractive effect in "undoped" lithium niobate. Peterson et al. (1971) and Clark et al. (1973) have suggested, though not conclusively shown, that the iron replaces a lithium ion in the crystal lattice. Recently, Keune et al. (1975) have suggested that the most likely site of the Fe³⁺ ion is the Nb site based on the Mössbauer-effect study of iron impurities in LiNbO₃. They were not able to identify the most likely site for the Fe²⁺ ions.

Conversion of iron impurities to the divalent state increases
the optical absorption in the region of 470 nm with a subsequent increase in the photorefractive sensitivity. It has been suggested (Clark et al. 1973) that optical absorption causes the intervalence transfer* of an electron from a Fe$^{2+}$ ion to a Nb$^{5+}$ ion. The Fe$^{2+}$ is converted to Fe$^{3+}$ when it loses an electron. After the Fe$^{2+}$→Nb$^{5+}$ transfer, the electron is free to move in the conduction band which is made up of Nb d orbitals. Retrapping occurs at Fe$^{3+}$ ions.

As might be expected, the photorefractive sensitivity may be controlled by the oxidation state of the iron impurities. Smith et al. (1968) found that field annealing lithium niobate at 600°C with a current density of 5 ma/cm$^2$ decreased the photorefractive sensitivity of the crystal. Later Peterson et al. (1971) showed that simply annealing the crystal in air or oxygen at 600°C for about 75 hours decreased the induced change about 22 times. Field annealing made any induced change undetectable. The applied field caused the iron ions to migrate towards the negative electrode. A yellow-brown deposit eventually appeared on the negative electrode as iron came right out of the crystal. Using EPR techniques, it was shown that annealing the crystals in air or oxygen converted about 96% of the iron to Fe$^{3+}$ (Clark et al. 1973).

Methods which convert Fe$^{3+}$ to Fe$^{2+}$ include heating the crystals in an argon atmosphere and heating the crystals in air while packed in a lithium salt such as Li$_2$CO$_3$ (Phillips and Staebler 1974a). The Li$_2$CO$_3$ treatment is discussed in Chapter 3.

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* A definition of intervalence transfer is given in Sec. 9.3 (Hush 1967).
2.9 Discussion

At the present time, it is thought that the charge transport involved in the photorefractive effect may be described by

\[ J = n \mu E + eD \frac{dn}{dx} + \kappa a I \]  

(2.30)

The relative contributions of the drift, diffusion and bulk photovoltaic terms are unsettled. These will be discussed in the following three sections.

The space charge field \((E_{sc})_j\) may be calculated using Eqs. 2.4, 2.6, 2.21 and 2.30 and the change in the indices of refraction found from

\[ \Delta n_i = -\frac{n_i^3}{2} r_{ij} (E_{sc})_j \]  

(2.31)

where \(r_{ij}\) is the appropriate electro-optic coefficient and \(n_i\) is the appropriate index of refraction.

2.9.1 Bulk Photovoltaic Effect

The new term in Eq. 2.30 \((\kappa a I)\) is formally equivalent to the current density which would be produced by drift in a constant field (either built-in or applied) provided that the light intensity did not change appreciably over distances comparable to that travelled by an electron before trapping. In this case, the extra electron concentration produced by light is \(g \tau\) where \(g\) is the local rate of generation of free electrons (proportional to \(\alpha I\)) and \(\tau\) is their lifetime. With a field \(E_v\), the drift current is \(eg \tau \mu E_v\). If we assume that the electrons do not in fact travel far in the above sense, we may allow for the new process by considering a "virtual" field to be added on to whatever fields are present, due to space charge or external application. The magnitude of \(E_v\) is uncertain and is probably dependent on various
properties of the crystal. In one case Glass et al. (1974b) claim that the photocurrent could be reduced to zero with an applied field of $6 \times 10^4$ V/cm for a radiant intensity of 0.32 W/cm$^2$ decreasing to $3.8 \times 10^4$ V/cm for 0.08 W/cm$^2$. In another case (Glass et al. 1975a) they show that $1.8 \times 10^3$ V/cm was required to cancel the photocurrent for 0.32 W/cm$^2$ and $1.2 \times 10^3$ V/cm for 0.08 W/cm$^2$. The experimental data in Chapter 8 suggests that the "virtual" field, $E_v$, is approximately $10^3$ V/cm in the crystal investigated. The analyses of hologram development using only drift and diffusion presented earlier in this chapter are not invalidated by the presence of the bulk photovoltaic effect. The extent to which the photovoltaic effect is responsible for the process depends on the magnitudes of other fields present. The experimental data in Chapter 8 demonstrates this point.

It should, however, be noted that the term $k_\text{AI}$ does not exactly describe the result of the proposed new mechanism when the light intensity changes rapidly with distance. Thus, the impulse response to a delta function of light $\delta(x - a)$ involves an exponential tail associated with the loss of the initial momentum, in addition to the effects of diffusion and drift which have been discussed previously. Although the mean distance involved in the initial flight is only 0.08 nm, according to Glass et al. (1974b), this distance is too short to be directly meaningful and must imply that most excited electrons do not escape from their traps. Those that do escape may travel quite appreciable distances on the scale defined by the hologram grating.
2.9.2 Built-in Field of Pyroelectric Origin

Although a new transport mechanism is believed to be involved, the presence of built-in fields of pyroelectric origin may also be important in some circumstances. As was discussed in Sec. 2.4 cooling the crystals from a higher temperature will create large fields. The fields on the surfaces of the crystal will normally disappear with the passage of time due to stray charges or leakage paths. However, the fields are so large that it seems quite possible that injection or extraction of electrons should set up space charges within the crystal with the result that large fields are still present in the crystal even after applying an external short.

2.9.3 Diffusion

As was discussed in Sec. 2.5.5, a diffusion-dominated process would require that the total field in the crystal be less than 100 V/cm. This would include the virtual field due to the bulk photovoltaic effect. Glass et al. (1974b) have used an argument similar to that discussed in Sec. 2.5.5 to claim that diffusion currents are negligible compared to photocurrents. Their evidence is that space charge fields of $10^5$ V/cm can be produced. However, as was discussed in Sec. 2.9.1, the development of space charge fields limited to $10^3$ V/cm to $2 \times 10^3$ V/cm appear possible. In this case diffusion would play a significant role.

Staebler et al. (1972b, 1974a) claim to have stored holograms by diffusion. Some of the confusion over the precise role of diffusion has arisen because holograms are not readily stored when the c-axis of the crystal is perpendicular to the plane of the two beams used to form the hologram. Examination of the electro-optic tensor of LiNbO$_3$
illustrates why holograms may not be stored in this configuration.

The equation for the optical indicatrix is

\[
\frac{1}{n_e^2} - \frac{1}{r_{22}E_2} + \frac{1}{r_{13}E_3}x_1^2 + \frac{1}{n_o^2} + \frac{1}{r_{22}E_2} + \frac{1}{r_{13}E_3}x_2^2
+ \frac{1}{n_e^2} + \frac{1}{r_{33}E_3}x_3^2 + 2(-r_{22}E_1)x_1x_2
+ 2(r_{51}E_2)x_2x_3 + 2(r_{51}E_1)x_3x_1 = 1 \tag{2.32}
\]

where \(E_1, E_2\) and \(E_3\) are the electric field components in the \(x_1, x_2\) and \(x_3\) directions respectively; \(n_o\) and \(n_e\) are the ordinary and extraordinary indices of refraction.

In the usual configuration for storing holograms, the \(c\)-axis \((x_3)\) is in the plane of incidence and normal to the bisector of the two beams as shown in Fig. 2.5(a). This creates an electric field \(E_3\).

From Eq. 3.32 the major effects are a change in \(n_e\) proportional to \(r_{33}\) \((30.8 \times 10^{-10} \text{ cm/V})\) and a change in \(n_o\) proportional to \(r_{13}\) \((8.6 \times 10^{-10} \text{ cm/V})\). The change in \(n_o\) occurs whether the light is propagating in the \(x_1\) or \(x_2\) directions.

If the crystal is turned through \(90^\circ\) so that the \(c\)-axis is normal to the plane of incidence, the field component that is created depends in which crystal direction the light is propagating. Fig. 2.5(b) shows the case where the grating vector (the direction normal to the planes of constant refractive index) is in the \(x_2\) direction. This creates a field \(E_2\). A change in \(n_o\) proportional to \(r_{22}\) \((3.4 \times 10^{-10} \text{ cm/V})\) will result. The only change in \(n_e\) will be caused by the cross term \(2r_{51}E_2x_2x_3\) which produces a rotation of the indicatrix. This however has a very small effect on \(n_e\). To read holograms in this con-
Fig. 2.5 Relations of the crystal axes and the two writing beams for different configurations of forming holograms.
figuration the electric vector of the light would have to be polarized in the $x_2$ direction. The efficiency would be about 100 times less than for holograms stored in the configuration of Fig. 2.5(a).

Fig. 2.5(c) shows the case where the grating vector is in the $x_1$ direction. This creates a field $E_1$. In this case there is no direct effect on either $n_e$ or $n_o$, the only change in indicatrix being a small rotation. It is evident that the $E_3$ component of the space charge field has the greatest effect on the indices of refraction. The fact that holograms are not easily stored when the $c$-axis of the crystal is normal to the planes of incidence is not a valid argument against diffusion as a transport mechanism. Amodei (1971b, 1972b) has shown that space charge fields of 1500 V/cm may be developed by diffusion. Chen (1969) reported storing holograms in the configuration of Fig. 2.5(b) at much reduced efficiency. (Chen did not specifically state the orientation except to say that the $c$-axis was normal to the plane of incidence.) Although Amodei believed diffusion to be a valid transport mechanism, he was unable to explain why he could not store holograms with the $c$-axis normal to the plane of incidence. In Chapter 8 more evidence is presented to support hologram formation both by diffusion and by the bulk photovoltaic effect.
CHAPTER 3

ELLIPSOMETRIC PROBE OF THE PHOTOREFRACTIVE EFFECT IN LiNbO$_3$

3.1 Introduction

A useful method for probing the photorefractive effect in LiNbO$_3$ involves the measurement of the birefringence of a sample at an array of positions before and after the optically-induced change in the indices. In this study an ellipsometer was used to measure the change in birefringence.

In ellipsometry (Fig. 3.1) the signal to the detector is nulled by rotating the analyser and polarizer with a quarter wave plate set at ±45° azimuth. The changes in relative phase and amplitude of two orthogonal linear polarizations are obtained as discussed in detail later in this chapter.

In the adjustable-compensator method used by Chen (1969) and others (Glass 1972, Serreze 1973) the fast and slow axes of a Soleil-Babinet compensator and the crystal coincide. The analyser and polarizer are crossed at ±45° azimuth and a null is obtained by adjusting the compensator phase retardation. This method does not allow for dichroism, while ellipsometry does.

Ellipsometry has been used extensively for the study of films on surfaces by reflected light (see Appendix G) but it does not seem to have been applied to the present problem apart from work in this laboratory (Wong 1973, Cornish, Moharam and Young 1975a). Automation is necessary to realize the potential of the method which would otherwise be excessively laborious. In our system, the instrument is balanced under computer control on a line or grid of points before and after
Fig. 3.1 Schematic of computer-controlled ellipsometer system.
exposure to the laser beam which causes the index changes. The optical properties which are probed using ellipsometric or adjustable-compensator techniques differ from those probed using holographic techniques. From ellipsometric measurements, the birefringence may be calculated. In holography, changes in the index for a particular polarization are observed rather than the birefringence.

3.2 Theory and Operation of the Ellipsometer

The ellipsometer measures the ratio ($\rho$) of the complex transmittivities for light with the E-vector along the c-axis ($T_\pi$) and at right angles to the c-axis ($T_\sigma$). When no multiple reflections of light occur within the sample,

$$\rho \equiv \tan \psi \exp \Delta = \frac{T_\pi}{T_\sigma} = \frac{t_{1\pi} t_{2\pi} \exp(-i\delta_\pi)}{t_{1\sigma} t_{2\sigma} \exp(-i\delta_\sigma)} \quad (3.1)$$

where $t_{1\pi}$ and $t_{2\pi}$ are the Fresnel transmission coefficients for the air/crystal and crystal/air interfaces $\pi$-light, similarly for $t_{1\sigma}$ and $t_{2\sigma}$; $\delta_\pi = 2\pi d n_e/\lambda_o$, where $d$ = sample thickness, $n_e$ = extraordinary index (as modified by the electro-optic effect), $\lambda_o$ = vacuum wavelength; and $\delta_\sigma$ is similarly defined using $n_o$, the ordinary index.

Light from a monochromatic, unpolarized source (He-Ne laser in this case) is passed through the polarizer which produces linearly polarized light of azimuth $P$. After passage through the quarter wave plate, elliptically polarized light is incident on the sample. The two orthogonally polarized waves, after passing through the sample, suffer a relative phase retardation ($\Delta$) and a relative amplitude reduction ($\tan \psi$).

For a measurement, the ellipsometer is balanced by varying
the parameters of the elliptic polarization of the light incident upon the sample until the transmitted polarization is linear and can be extinguished when the analyser is set to azimuth A. The parameter variation is achieved by rotating the polarizer with the quarter wave plate fixed at ±45° azimuth. (For all measurements in this thesis, the QWP = -45°.) This method of varying the ellipticity of the light is chosen because the calculation of Δ and tan ψ from the extinction azimuths of P and A is greatly simplified.

The convention for measuring angles is as follows. For the normal mode of operation, that is for measurements made by reflecting light from the sample, the polarizer, analyser and quarter wave plate angles are measured from the plane of incidence. Positive angles are measured counter-clockwise when looking towards the light source. In the experiments to be described in this chapter, the light from the polarizer arm of the ellipsometer was normally incident on the crystal. The optical properties of the crystal were probed by transmitting light through the crystal rather than reflecting light from its surface. The c-axis of the crystal was aligned to be perpendicular to the normally incident beam and at zero azimuth.

In determining the relationships between Δ and P, and tan ψ and A it is convenient to follow the path of the light which is incident on the polarizer through to the analyser using the Jones vectors and matrices (Shurcliff 1962). The polarizer initially is at azimuth P and the fast axis of the quarter wave plate (QWP) is at Q. Referring to Fig. 3.2, the light from the polarizer, resolved in the directions of the fast and slow axes of the QWP can be represented by the normalized Jones vector.
The ratio of the complex transmittivities of the QWP may be written as

\[ \rho_c = T_c \exp(-i\Delta_c) \]
\[ = T_c (\cos\Delta_c - i\sin\Delta_c) \] (3.3)

where \( T_c \) is the ratio of the transmittance along the fast axis to that along the slow axis and \( \Delta_c \) is the relative phase retardation. The Jones matrix is
To rotate the coordinate axis of the light from the fast and slow axes of the QWP to axes parallel to the crystal's optic axis and perpendicular to this axis, the counter-rotator matrix

\[
\begin{bmatrix}
\cos Q & -\sin Q \\
\sin Q & \cos Q
\end{bmatrix}
\]  \hspace{1cm} (3.5)

is used. The sample is simply another birefringent plate and can be represented in a manner similar to the QWP, so that

\[
\rho_s = T_s \exp(i\Delta_s) = T_s (\cos \Delta_s + i\sin \Delta_s)
\]  \hspace{1cm} (3.6)

and the Jones matrix is given by

\[
\begin{bmatrix}
1 & 0 \\
0 & \rho_s
\end{bmatrix}
\]  \hspace{1cm} (3.7)

The state of polarization of the light after passing through the sample is given by

\[
\begin{bmatrix}
1 & 0 \\
0 & \rho_s
\end{bmatrix}
\begin{bmatrix}
\cos Q & -\sin Q \\
\sin Q & \cos Q
\end{bmatrix}
\begin{bmatrix}
1 & 0 \\
0 & \rho_c
\end{bmatrix}
\begin{bmatrix}
\cos(P-Q) \\
\sin(P-Q)
\end{bmatrix}
\]

\[
= \begin{bmatrix}
\cos Q \cos(P-Q) - \rho_s \sin Q \sin(P-Q) \\
\rho_s \{\sin Q \cos(P-Q) + \rho_c \cos Q \sin(P-Q)\}
\end{bmatrix}
\]  \hspace{1cm} (3.8)

For this light to be extinguished by the analyzer it must be linearly polarized with an azimuth \(\varepsilon\), given by

\[
\tan \varepsilon = \frac{\rho_s \{\sin Q \cos(P-Q) + \rho_c \cos Q \sin(P-Q)\}}{\cos Q \cos(P-Q) - \rho_c \sin Q \sin(P-Q)}
\]
\[ \frac{\rho_s \{\tan Q + \rho_c \tan(P-Q)\}}{1 - \rho_c \tan Q \tan(P-Q)} = -\tan^2 \phi. \] (3.9)

The analyser must be 90° from e to extinguish the light, so that

\[ \tan A = \frac{-1}{\tan e} \]

\[ = \frac{\rho_c \tan Q \tan(P-Q) - 1}{\rho_s \{\tan Q + \rho_c \tan(P-Q)\}}. \] (3.10)

Assuming a perfect quarter wave plate \((T_c = 1)\), with \(Q = 45°\), \(\rho_c = -i\) and \(\tan Q = -1\). Eq. 3.10 can be solved for \(\rho_s\) to give

\[ \rho_s = \frac{1 - i \tan(P+45°)}{\tan A \left[1 + i \tan(P+45°)\right]} \] (3.11)

Substituting Eq. 3.6 and equating real and imaginary parts yields

\[ \Delta_s = -2P - 90° \pm n\pi, \quad n = 0,1,2,... \] (3.12)

and

\[ T_s = \pm 1/\tan A \]. \] (3.13)

Thus the polarizer reading is proportional to the phase change and the tangent of the analyser reading is proportional to the inverse of the transmittance ratio. The relation between the phase change and the birefringence of the crystal is

\[ \Delta_s = \frac{2\pi d}{\lambda} (n_e - n_o). \] (3.14)

\(T_s\) is related to the ratio of the absorption coefficients along the fast and slow axes of the crystal. For constant thickness, a change in \(P\) indicates a change in birefringence and a change in \(A\) indicates a change in the ratio of the two absorption coefficients along the two principal directions.
3.3 Sensitivity of the Ellipsometer

Using Eqs. 3.12 and 3.14 the sensitivity of the ellipsometer to changes in birefringence can easily be computed. If $P_i$ is the initial polarizer reading and $P_f$ the reading after a change in birefringence $\Delta(n_e - n_o)$, then

$$\Delta(n_e - n_o) = \frac{(P_i - P_f) \lambda}{\pi d}$$

(3.15)

The sensitivity of the polarizer (and analyser) reading is $0.01^\circ$ corresponding to a change of $3.5 \times 10^{-8}$ in birefringence for a 1.0 mm thick crystal.

However, the resolution of the measurements is limited by the temperature dependence of the refractive indices. For a $1^\circ$C change in temperature, the change in the birefringence (at $\lambda = 650$ nm) is $\approx 0.5 \times 10^{-4}$ (Boyd et al. 1967). For a 1.0 mm thick crystal this corresponds to $13^\circ$ change in the polarizer.

Errors may arise in ellipsometry due to imperfect components and imperfect alignment of the instrument. These errors however, are negligible compared to the uncertainty introduced by small fluctuations in the temperature of the crystal. (See Appendix F.)

3.4 The Automated Ellipsometer

The ellipsometer was a Rudolph type 43603-200E, modified for computer control (PDP 8/E) as indicated in Fig. 3.1. A Spectra Physics 1 mw He-Ne laser (model 133, unpolarized) was used as a light source. The detector was a photomultiplier (RCA 8645 tube with a Kepco regulated voltage source). It was mounted on the end of the analyser arm with a pin hole, a ground glass diffuser and a 632.8 nm interference filter
(10 nm band pass) used to restrict spurious light signals from illuminating the detector. This allowed the instrument to be used with normal room lighting. The Rudolph 546.1 nm quarter wave plate was replaced with a quartz Soleil-Babinet compensator (Gaertner model L-135) mounted on the Rudolph graduated circle and set for quarter wave retardation at 632.8 nm.

The polarizer and analyser were driven through anti-backlash gears by stepping motors (IMC Magnetics Corp. #PIN 008-008) with absolute shaft encoders (Decitrak TR 511-CW/D) to read the angles. The sensitivity of the shaft encoders was 0.01° which was one motor step. Balances reproduced with little more scatter. The encoder output unit converted the angles to binary-coded decimal for input to the computer via the interface. The interface was constructed mostly of standard Digital Equipment Corp. components. The motor drives were controlled through the interface. The error signal from the detector was amplified by a variable gain amplifier and interfaced with the computer through an analog-to-digital converter (DEC A811, accuracy of 0.1% F.S.). The analog error signal was displayed on a meter to allow manual nulling of the ellipsometer.

A second set of stepping motors was used to move the crystal about in a plane perpendicular to the polarizer arm of the ellipsometer. Anti-backlash gears between the motor and stage allowed the sample to be moved in 0.8 μm steps, if desired.

Programs and data were stored on a dual Dectape unit. An incremental ploter (Houston model DP-10) allowed graphical output. The programs to control the system were written in a combination of Fortran and assembly language and are listed in Appendix H.
The balance procedure used was based on the principle that the light at the detector varies symmetrically for small excursions of the polarizer and analyser from their balance positions (Archer 1962). The polarizer was balanced first and then the analyser. This was repeated at each balance to reduce scatter. The computer determines which way it must drive the polarizer to reduce the error signal and it drives the motor until the error signal goes through the minimum. It then sums a number of readings (usually 12) after each step and then reverses, driving the motor through the position of minimum signal. It then takes a running sum of readings on the present side of the minimum, adding one and dropping the twelfth previous reading, until this sum equals the sum taken on the other side of the minimum. The balance point (the mid point between the sums) is calculated and the polarizer driven to that point. The analyser is then balanced in the same manner. This entire procedure is completed a second time and then the analyser and polarizer readings, as given by the shaft encoders, are stored.

To measure the birefringence along, for instance, the c-axis of the crystal, the ellipsometer is balanced and then the crystal is moved a small distance perpendicular to the probing beam. This procedure is followed repeatedly and after each ellipsometer balance, the analyser reading, polarizer reading and crystal position are stored in the computer memory. When the memory buffer is filled the data is transferred to the magnetic tape unit and the process continues to completion. During the scan the polarizer reading may be plotted. After the scan, the data stored on the magnetic tape may be processed and plotted.
3.5 Sample Alignment

The crystals were aligned using the He-Ne laser of the ellipsometer. The crystals were supplied (see Appendix D) with the edges of the rectangular crystals parallel to the crystal axes. The c-axis of the crystal was aligned to zero azimuth by adjusting the crystal tilt to make the appropriate edge parallel to the laser beam. To adjust the angle of incidence of the laser beam to 90°, the crystal was adjusted to cause the beam to be reflected back on itself. The crystal was then slightly misaligned so that the reflected beam would not enter the pin hole of the quarter wave plate. This prevented the possibility of problems due to multiple reflections between the sample and the quarter wave plate (Oldham 1967).

3.6 Temperature Control for Ellipsometer Measurements

As was mentioned previously, thermostating the crystal is necessary for measurements made on the ellipsometer. Fig. 3.3 shows a schematic of the apparatus used. The crystal was enclosed in an insulated plastic box. A fan and a proportional temperature controller (YSI model 72) with a thermister detector and a fine wire heater were used to maintain the temperature to within ±0.02°C. No windows were used, the ellipsometer arms projecting through holes in the box. A digital thermometer (HP model 2802A) was used to measure the absolute temperature to approximately ±0.05°C, and another thermister in a bridge circuit was used to measure variations in temperature with a sensitivity of 0.01°C. The temperature was maintained a few degrees above room temperature (approximately 29°C). Fig. 3.4 gives an indication of the temperature stability achieved. Since most of the scatter in ellipso-
Fig. 3.3 Schematic of the apparatus used to thermostat the lithium niobate crystals during ellipsometer measurements.

Fig. 3.4 Typical measurement of the temperature stability inside the insulated box. The temperature was monitored with the thermister bridge circuit.
meter readings was due to temperature fluctuations, better control would extend the sensitivity of the instrument. However the setup was adequate for the present purpose. The effectiveness of temperature control increases if the optical pathlength is reduced so that thin crystals are more suitable for this type of experiment.

Fig. 3.5 shows the effect of repeatedly balancing the ellipsometer with the LiNbO₃ crystal (3mm thick) fixed at one position, while the temperature slowly changed from 28.0°C to 24.0°C.

Fig. 3.6 is typical of the scatter in the polarizer readings. The points are the difference in the polarizer readings for two repeated scans along the c-axis of a crystal (3 mm thick).

3.7 The Effects of Multiple Internal Reflections

3.7.1 The Effect on the Measurement of the Birefringence

When multiple reflections of the light occur between the surfaces of the crystal, the relations of the ellipsometry readings to the optical properties of the crystal are not as straightforward as previously shown. If the light source employed is a laser, the coherence length will generally be longer than a few centimeters and therefore always longer than the thickness of the crystals examined (which are <1 cm). In considering the passage of an optical wave through a crystal, both the amplitude and phase must be accounted for. Because a portion of the wave will be reflected each time it is incident on a boundary, many waves will be present within the sample, half travelling in a direction other than that of the incident light wave. Constructive and destructive interference will occur. The situation is analogous to the transmission of light through thin solid films.
Fig. 3.5 The effect of temperature on the polarizer reading when measuring the birefringence in a 3 mm thick crystal of LiNbO$_3$.

Fig. 3.6 The difference in the polarizer readings of two scans along the c-axis showing the scatter in the readings.
For the case where there are no multiple reflections, Eq. 3.1 holds. However, when multiple reflections are considered the expressions for the transmittivities, $T_{\pi}$ and $T_{\sigma}$, are of the form

$$T = \frac{t_1 t_2 \exp(-i\delta)}{1 - r^2 \exp(-2i\delta)} .$$

(3.16)

Substituting these in Eq. 3.1, the result obtained is

$$\rho = \frac{A (1 - r_{\sigma}^2 \exp(-2i\delta_{\sigma}))}{(1 - r_{\pi}^2 \exp(-2i\delta_{\pi}))}$$

(3.17)

where $A$ is the value $\rho$ would have if multiple reflections are absent. Here $r_{\sigma}$ and $r_{\pi}$ are Fresnel reflection coefficients. From this formula, it follows that the observed $\Delta$ and the relative phase change ($\delta_{\pi} - \delta_{\sigma}$) are related through

$$\tan \Delta = B \tan(\delta_{\pi} - \delta_{\sigma}),$$

(3.18)

where

$$B = \frac{1 - R_{\sigma} R_{\pi} + (R_{\sigma} - R_{\pi}) \sin(\delta_{\sigma} + \delta_{\pi})/\sin(\delta_{\sigma} - \delta_{\pi})}{1 + R_{\sigma} R_{\pi} - (R_{\sigma} + R_{\pi}) \cos(\delta_{\sigma} + \delta_{\pi})/\cos(\delta_{\sigma} - \delta_{\pi})}$$

and $R_{\sigma} = r_{\sigma}^2$, $R_{\pi} = r_{\pi}^2$. Here $B$ would be unity if multiple reflections were absent. The importance of multiple reflections does not seem to have been noted when using this method (or the adjustable-compensator method). Using these methods, data that has been previously published may be in error to some extent (Chen 1969, Serreze et al. 1973, Glass et al. 1972, Glass et al. 1974b, Wong 1973). Fig. 3.7 shows the change in $\Delta$ (vertical scale) as a function of change in birefringence and also of thickness for assumed values of the various parameters. (Here $n_o$ and $n_e$ were assumed to have changed due to a space charge field, with}
Fig. 3.7 Calculated change in $\Delta$ (vertical) as a function of (a) change in birefringence $\Delta(n_e - n_o)$ (calculated appropriate to electro-optic effect with field as shown) and (b) thickness. Probe wavelength = 632.8nm.
the numerical values of the electro-optic coefficients from Turner (1966) being used.)

For the range shown, the change in $\Delta$ is nearly proportional to the change in the birefringence, but the sensitivity depends on the optical thickness of the crystal.

3.7.2 The Effect of Multiple Internal Reflections on the Photorefractive Process

The photorefractive process in lithium niobate depends on the intensity of the illumination and, in the initial stage of the process, the relationship is believed to be linear. Insight into the effect of multiple reflections can be gained by considering the simple case of a plane, uniform light wave normally incident on the crystal. In the absence of multiple reflections the intensity will be uniform throughout the crystal (no absorption). With multiple reflections, the amplitude $f(z)$ of the electric field as a function of distance $z$ into the crystal may be obtained by summing beams. Referring to Fig. 3.8 the sum of the beams at a distance $z$ into the sample is

$$f(z) = t_1 \exp(-i2\pi nz/\lambda) + t_1r_2 \exp(-i2\pi n(2d - z)/\lambda) + t_1r_2r'_1 \exp(-i2\pi n(2d + z)/\lambda) + \ldots$$

(3.19)

This is a geometric series with a sum

$$f(z) = \frac{t_1 \exp(-iz\delta/d)(1 + r \exp(-2i(1 - z/d)\delta))}{1 - r^2 \exp(-2i\delta)}$$

(3.20)

where $r_2 = r'_1 = r$, the Fresnel reflection coefficient of the crystal/air interface and $\delta = 2\pi nd/\lambda$ for normal incidence.
Effectively, a standing wave is set up between the crystal surfaces, and the amplitude of the wave varies periodically through the crystal. If the crystal thickness is changed slightly, the relative phase of the waves launched after reflection will be different, and the interference of the forward travelling and backward travelling waves will be altered. If the mean intensity of the light within the sample is calculated, it is found to vary periodically with sample thickness as shown in Fig. 3.9.

Because $n_e$ and $n_0$ are not equal, different mean intensities within the crystal will result for $\pi$ and $\sigma$ polarized light for the same incident intensity.

3.8 Birefringence Measurements Along the c-axis of the Crystal

In initially probing the crystals along the c-axis it was found that the polarizer reading varied considerably. Figs. 3.10 and 3.11 show scans along two undoped LiNbO$_3$ crystals and Fig. 3.12 shows a scan along an Fe-doped (0.015 mole%) crystal.

Considering Fig. 3.11, it would appear that either the
Fig. 3.9 Variation with thickness in the mean intensity of light, due to multiple reflections. The incident beam is of unit intensity. \(d = \text{thickness, } d_o = 1 \text{ mm and } \lambda = 441.6 \text{ nm.}\)
Fig. 3.10 The variation in the polarizer reading along the c-axis of the undoped crystal #5 (see Appendix D).
Fig. 3.11 Variation in the polarizer (P) and analyser (A) readings along the c-axis of the undoped crystal #4 (see Appendix D).
Fig. 3.12 Variation in the polarizer reading along the c-axis of the Fe-doped crystal #2 (see Appendix D).
birefringence or the thickness are varying periodically along the c-axis (variation in P) and also that the dichroism is varying periodically (variation in A). However, when multiple reflections are allowed for the data in Fig. 3.11 can be fitted, using Eq. 3.17, assuming a simple gradient in thickness and extraordinary index, with the ordinary index remaining constant. This is shown in Fig. 3.13. The gradient in thickness is within the tolerances of the optical polishing (sides parallel to 10 arc seconds) and the gradient in \( n_e \) could have been caused by a gradient in the non-stoichiometry of the sample as it was pulled from the melt. It is believed that \( n_e \) is dependant on the Li\(_2\)O:Nb\(_2\)O\(_5\) ratio but that \( n_0 \) is not (Bergman et al. 1968).

3.9 Optically-Induced Birefringence Change due to a One-Dimensional Gaussian Beam

3.9.1 Introduction

Wong (1973) has shown that considerable simplification of the analysis of the optically-induced index change is achieved when the problem is made spatially one-dimensional, and exposure is minimized to reduce saturation effects.

To investigate the photorefractive effect, crystals were irradiated with a narrow strip of light from a He-Cd laser (\( \lambda = 441.6\)nm) that spanned the crystal as shown in Fig. 3.14. The diameter of the laser beam was reduced with two microscope objectives and then expanded with a cylindrical lens to give a gaussian profile along the c-axis, as shown in Fig. 3.15. The curve in Fig. 3.15 was measured with a Gamma Scientific Model 2900 scanning auto-photometer.
Fig. 3.13 The circles are the polarizer readings shown in Fig. 3.11. The solid curve was calculated with the following assumed parameters: thickness = 1.00 mm + 1.92 wavelengths/cm, $n_e = 2.20657 + 3.75 \times 10^{-5}$/cm, $n_o = 2.29058$. 
Fig. 3.14 Method used to illuminate crystals with a narrow beam of light.

Fig. 3.15 Profile of the light intensity along the c-axis of the crystal for the method of illumination shown in Fig. 3.14.
3.9.2 Theoretical Considerations

The shape of the expected birefringence can be determined mathematically for a simplified model. As will be shown, the ellipsometer can be used to differentiate between induced index changes due to a drift dominated process and a diffusion dominated process. The migration length of the excited electrons can be measured if it is long enough to be resolved by the instrument.

For the linearized model where the space charge field is small compared to the total field in the crystal and the trap occupancy is considered to be only slightly perturbed, the rate of generation of electrons is proportional to the light intensity, and the rate of capture of electrons is proportional to the free electron concentration. This is analogous to the case discussed in Chapter 2 except that the spatial variation of the light intensity is gaussian rather than sinusoidal. The equations of Chapter 2 which govern the process are

\[ J = -ne\mu E_0 + eD\partial n/\partial x \]  
\[ \frac{\partial n}{\partial t} = -\frac{n}{\tau} + \frac{1}{e} \frac{\partial J}{\partial x} + g(\text{light}) = 0 \]  
\[ \frac{\partial \rho}{\partial t} = -\frac{\partial J}{\partial x} \]  
\[ \frac{\partial E_{sc}}{\partial x} = \frac{\rho}{\varepsilon} . \]

The current due to the photovoltaic effect is formally equivalent to what would be produced by drift in an electric field, as discussed in Chapter 2. This effect is included as part of \( E_0 \). For the drift-only case, the space charge field is given by

\[ E_{sc}(x) = n(x)eE_0\mu_n / \varepsilon . \]
For the diffusion-only case, the space charge field is given by

\[ E_{sc}(x) = -(eD't/e) \frac{\partial n(x)}{\partial x}. \]  (3.26)

Eq. 3.21 and 3.22 are used to calculate \( n(x) \) and \( dn/dx \). For arbitrary intensity variation, the number of electrons in the conduction band, \( n(x) \), is given by the convolution of the intensity pattern, \( g(x) \), with the impulse response, \( h(x) \), at \( x = 0 \), for the process being considered (Wong 1973, Young et al. 1974).

For drift, the impulse response is

\[ h(x) = \frac{u(x)}{\mu E_0} \exp(-x/L) \]  (3.27)

where \( u(x) \) is the unit step function, \( L \) is the drift length and is equal to \( \mu E_0 \tau \). For diffusion, the impulse response at \( x = 0 \) is two decaying exponentials back-to-back:

\[ h(x) = \frac{1}{L'} \exp(-|x|/L') \]  (3.28)

where \( L' = (D\tau)^{1/2} \) is the diffusion length. The convolution of \( g(\text{light}) \) with the impulse response is

\[ g(x) \ast h(x) = \int_0^\infty g(y)h(x-y)dy \]  (3.29)

where the gaussian light intensity function has the form

\[ g = g_o \exp\left(-a^2x^2\right). \]

Fig. 3.16 shows the spatial variation of \( g(x) \), \( n(x) \) and \( E_{sc}(x) \) for the two cases of drift and diffusion. The situation illustrated is for long migration length. If the migration length were very short, \( n(x) \) would be a replica of \( g(x) \).
Fig. 3.16 The space charge fields ($E(x)$) developed for the cases of drift and diffusion, caused by the intensity distribution $g(x)$. $n(x)$ is the spatial distribution of electrons in the conduction band during illumination.
3.9.3 Experimental Results

The results of measurements on a number of crystals, both undoped and Fe-doped (0.015 mole %) all showed the shape characteristic of a drift-dominated process. Fig. 3.17 illustrates a typical measurement on an undoped crystal. There was no apparent one-sided exponential tail on the curves. In the doped crystals, the magnitude of the induced index change was as much as ten times as great as that seen in undoped crystals.

Fig. 3.18 shows the results of a measurement on an Fe-doped crystal. Instead of a single peak occurring as would be expected, two peaks developed. Possibly, the large change in the indices was sufficient to appreciably change the optical path length of the crystal. Due to multiple reflections, the sensitivity of the ellipsometer may have been decreased (see Fig. 3.7) causing $\Delta P$ to decrease in the region between the two peaks even though $\Delta (n_e - n_o)$ may have increased.

In the narrow beam experiments, the shape of the curve is what would be expected for a process involving drift rather than one involving diffusion. The lack of an exponential tail on either side of the illuminated region indicates that the migration length is smaller than the resolution of the instrument ($<<100\mu m$). The spatial resolution is limited by the diameter of the probing beam since, in the setup used, the sample could be displaced in steps of approximately $l\mu m$. The resolution might be increased by reducing the probing beam diameter and using deconvolution techniques to extract the information on the spatial variation in the birefringence. This would be useful for examining materials in which the electrons were displaced more than $1\mu m$.

Recently von der Linde et al. (1975a, 1975b) have claimed that photorefractive
Fig. 3.17 Ellipsometric scan of the optically-induced birefringence change caused by a single laser beam focussed by a cylindrical lens. **Top:** Change in polarizer reading $\Delta P$ along c-axis. **Bottom:** Scan of intensity across laser beam on same horizontal scale as $\Delta P$ curve.
Fig. 3.18 Ellipsometric scan of the optically-induced birefringence change in an Fe-doped (0.015 mole %) crystal. Top: Change in the polarizer reading along the c-axis. Bottom: Scan of the intensity of the laser beam causing the change.
processes in KTN involve the redistribution of electrons with a drift length of about 12 \( \mu \text{m} \).

3.9.4 Discussion

Ellipsometry is better suited than adjustable-compensator methods for studying photorefractive effects because in ellipsometry dichroism is accounted for while in adjustable-compensator methods it is not. In the adjustable-compensator method, in the absence of multiple reflections, fixed dichroism would result in a fixed error in estimating \( \Delta \), which would tend to cancel in examining changes due to the photorefractive effect. A change in dichroism due to the photorefractive effect would produce an error. Multiple reflections will lead to further errors in determining \( \Delta \) (quite apart from the problem of interpreting \( \Delta \)).

From Fig. 3.7 it can be seen that for a crystal of constant thickness the change in \( \Delta \) is nearly linear with the change in birefringence. Difficulties in interpreting \( \Delta \) are due to the variation of the sensitivity of the ellipsometer with the thickness of the sample at the point being probed.

3.10 Measurements on Crystals Heated in \( \text{Li}_2\text{CO}_3 \)

3.10.1 Introduction

Measurements were made on crystals before and after heating in \( \text{Li}_2\text{CO}_3 \) to investigate the effects of the treatment on the birefringence and on the photorefractive process. Previously, some effects of the treatment on hologram writing and on the optical absorption had been reported (Staebler et al. 1974, Phillips et al. 1974) which indicated that the treatment reduced iron ions from \( \text{Fe}^{3+} \) to \( \text{Fe}^{2+} \). The experiments
to be described indicate that the $\text{Li}_2\text{CO}_3$ treatment affects the birefringence of the crystal by decreasing the value of the extraordinary index. It is postulated that this is due to in-diffusion of $\text{Li}_2\text{O}$ into the crystal, which, in addition to causing iron reduction, serves to destroy shallow traps.

3.10.2 Experimental Procedures and Results

An undoped crystal grown from a stoichiometric melt was heated in air to 520°C for 40 hours while packed in $\text{Li}_2\text{CO}_3$ powder. This treatment is claimed to convert more than 96% of the iron impurities to the divalent state (Phillips et al. 1974). The crystal was scanned along the c-axis after the $\text{Li}_2\text{CO}_3$ treatment. The variation in the polarizer reading is shown by the lower curve in Fig. 3.19. The upper curve is a reproduction of Fig. 3.11 which shows the polarizer variation before the treatment. The periodic nature of the readings has been removed and the curve has shifted downwards. An attempt to fit the lower curve in Fig. 3.19 is shown in Fig. 3.20. The gradient in $n_e$ and the nominal value of $n_e$ were smaller than the values used to fit the curve measured prior to the $\text{Li}_2\text{CO}_3$ treatment. A better fit could probably be achieved by assuming a nonlinear change in $n_e$ along the sample. This would be consistent with uneven diffusion of lithium into the sample along the c-axis. When more heavily doped samples are heated in $\text{Li}_2\text{CO}_3$ this uneven diffusion is readily seen. The treatment causes the samples to turn brown and the effect is more pronounced near the unpolished edges of the sample.

Optical damage due to a narrow slit produced patterns of the same shape as were found for the untreated sample. However, the damage at different places along the c-axis was more uniform after treatment.
Fig. 3.19 The change in the polarizer reading (crystal #4) caused by heating in Li₂CO₃. TOP: before heat treatment (same as Fig. 3.11). BOTTOM: after heat treatment.

Fig. 3.20 The circles are the polarizer readings shown by the bottom curve in Fig. 3.19. The solid curve was calculated with the following assumed parameters: thickness = (1.0 mm - 0.19 wavelengths) + 1.92 wavelengths/cm; $n_e = 2.20642 + 1.25 \times 10^{-5}$/cm; $n_o = 2.29058$. (compare with Fig. 3.13).
Fig. 3.21(a) shows the change observed in polarizer readings due to optical damage produced by identical exposures at various places along the c-axis before treatment. This variability was removed by the Li$_2$CO$_3$ treatment as shown in Fig. 3.21(b). The variation is believed to indicate the combined effect of multiple reflections and the gradient in stoichiometry.

Thermal decay of the optical damage in undoped LiNbO$_3$ before and after Li$_2$CO$_3$ treatment was measured by taking repeated scans along the c-axis over a period of a few days. Fig. 3.22 shows how the change in the polarizer reading $\Delta P$ varied with time. The elapsed time from the end of the exposure creating the damage to the measurement of the birefringence at the peak in $\Delta P$ is given beside the curves. The curves do not all have the same baseline as shown. (This was normalized to illustrate the decay more readily.) The small shifts ($P < 1^\circ$) were attributed to the temperature not being exactly the same when each scan was taken. Effectively, a small increase in temperature lowers the polarizer readings.

In Fig. 3.23 the logarithm of the change in $\Delta P$ is plotted against time and shows an exponential decay with a time constant of approximately 46 hours. The same experiment was repeated for the same undoped crystal after the Li$_2$CO$_3$ treatment. After 30 hours, negligible decay had occurred.

3.10.3 Discussion

The Li$_2$CO$_3$ treatment introduced by Phillips and Staebler affects not only the absorption of LiNbO$_3$ but also the birefringence. In addition, thermal decay of the optical damage was diminished after
Fig. 3.21 Change in polarizer reading due to irradiation of three places along the c-axis with a one-dimensional Gaussian beam giving an average of 110 J/cm² at a wavelength of 441.6 nm. (a) before and (b) after lithium carbonate treatment.
Fig. 3.22 Thermal decay of optical damage in an undoped lithium niobate crystal before heating in Li$_2$CO$_3$. The times indicate the elapsed time from the end of illumination to the measurement of the peak in each curve (±1 min).
Fig. 3.23 Logarithm of the change in ΔP as a function of time. The experimental points correspond to the peak amplitudes of the curves in Fig. 3.22.
treatment. A decrease in the concentration of empty traps (Fe$^{3+}$) will increase the migration (or diffusion) length of electrons and will, therefore, enhance hologram writing, optical erasure and thermal decay. This is easily seen by noting that the farther electrons move, the greater the effect in all three cases. The diminished thermal decay seen cannot, therefore, be due to the reduction of iron centres.

If, however, the Li$_2$CO$_3$ treatment served to destroy shallow traps as well as to reduce iron centres to the Fe$^{2+}$ state, this would account for all the above observations. Electrons would be captured by the shallow traps during hologram writing and would contribute to the hologram (or optical inhomogeneity), but they would escape more readily by thermal activation than those trapped in the iron centres. Destruction of the shallow traps would diminish thermal decay. Since it would increase the distance travelled by free electrons before trapping (as would also occur with the reduction of iron centres) it would therefore aid in increasing optical writing and erasure. This explanation is consistent with previous reports (e.g. Chen et al. 1968) that thermal decay occurs in two stages, an initial rapid decay followed by a slower decay.

As to why treatment in Li$_2$CO$_3$ should produce these effects it is well known that out-diffusion of lithium (Kaminow et al. 1973) can be used to produce optical wave guides. LiNbO$_3$ can crystallize in a non-stoichiometric form, (Li$_2$O)$_v$(Nb$_2$O$_5$)$_{1-v}$, where $v$ ranges from 0.48 to 0.50 moles (Carruthers et al. 1971). It is believed that the ordinary index, $n_o$, is independent of $v$, but within the given range the extraordinary index, $n_e$, increases almost linearly as $v$ decreases (Bergman et
al. 1968). In the production of optical wave guiding layers, it was suggested (Kaminow et al. 1973) that Li$_2$O was released when the crystal was heated in vacuum, thus increasing $n_e$.

When LiNbO$_3$ is heated while packed in Li$_2$CO$_3$, it seems reasonable to postulate that Li$_2$O is diffused into the crystal. Introduction of O$^{2-}$ would destroy oxygen vacancies. If the oxidation state of the iron centres serves to maintain electroneutrality then introduction of extra Li$^+$ would explain the reduction of iron centres. The introduction of Li$_2$O is consistent with the decrease of $n_e$ required to fit the data of Fig. 3.19. No explanation for the reduction of the iron centres was given by Phillips and Staebler (1974a) but it is understood (personal communication) that they had also considered the above explanation.
CHAPTER 4

THE USE OF FABRY-PEROT FRINGES TO OBSERVE THE PHOTOREFRACTIVE EFFECT

4.1 Introduction

Multiple internal reflections of a laser beam between the two optically polished surfaces of a crystal produce light and dark fringes, depending on the optical thickness of the crystal. The situation is similar to that found in a Fabry-Perot etalon (Born and Wolf 1959). The optical path change required to move a dark fringe to the position of the next adjacent dark fringe is $\lambda/2$ where $\lambda$ is the wavelength of illumination.

The geometry of the fringes is an indication of the variation of the optical thickness of the crystal. Optical inhomogeneities can easily be seen with this technique. The fringes can be made to move by heating the sample, or by optically-induced refractive index changes. Changes in the ordinary and extraordinary indices can be viewed independently and the magnitude of the optical change estimated. This technique is especially useful for visual inspection of the spatial variation of optically-induced inhomogeneities.

4.2 Experimental Procedures

The fringes are easily visible to the eye when the reflection of an expanded laser beam from the surface of the crystal is viewed. The optical arrangement for the photographs taken in this Chapter is shown in Figure 4.1.
Fig. 4.1 Optical arrangement for taking photographs of the Fabry-Perot interference fringes.
The beam from a laser (either He-Ne or argon ion) was spatially filtered and expanded to illuminate the entire crystal. A beam splitter was placed before the crystal to deflect the reflected beam towards the camera.

4.3 Experimental Results

Photographs of the Fabry-Perot fringes are shown in Fig. 4.2. For Fig 4.2(a) - 4.2(d), an oscilloscope camera was used with a diffuser placed where the scope face would usually be. The crystal was illuminated with an argon ion laser ($\lambda = 541.5\text{nm}$). The two photos of Figs. 4.2(e) and 4.2(f) were taken with a 35mm camera without a lens. The light source was a He-Ne laser ($\lambda = 632.8\text{nm}$).

The photos on the left (Fig. 4.2(a), 4.2(c), 4.2(e)) correspond to the extraordinary index and those on the right (Fig. 4.2(b), 4.2(d), 4.2(f)) to the ordinary index. Fig. 4.2(a) and 4.2(b) show the crystal after any optically-induced inhomogeneity has been annealed out of the crystal by heating it for a few hours at 270°C. The vertical band towards the right end of the photos is partially due to faulty polishing of the surface and partially due to optical inhomogeneities in the bulk produced during growth.

The photos in Fig. 4.2(c) and 4.2(d) show two effects: The thin vertical line on the left is the result of irradiating the crystal with a narrow strip of light as described in Chapter 3. The spatial extent of the index change is approximately as wide as the beam. In the central area of the crystal, the fringes have been heavily distorted after a number of holograms have been stored in the crystal. These effects are due to the build-up of large dc space charge fields within the
Fig. 4.2. Fabry-Perot fringes in an Fe-doped crystal showing optically-induced changes in the refractive indices. (a), (c), (e) were made with extraordinary polarized light and show the variation in $n_e$. (b), (d), (f) were made with ordinary polarized light and show the variation in $n_o$. 
crystal. The sinusoidal variations in index that produce diffraction would be too small to be seen.

Fig. 4.2(e) and 4.2(f) show optical damage due to irradiation with a single beam of circular symmetry. These patterns are analogous to the experimental results of Chen (1969) except that here the variation in \( n_e \) and \( n_o \) are seen separately rather than the variation in the birefringence. The damaged portion shown in Fig. 4.2(f) was approximately the same size as the beam. The damage in Fig. 4.2(e) extends well beyond the edges of the light beam. This is in contrast to the thin line shown in Fig. 4.2(c) and 4.2(d).

Damage is more apparent for the extraordinary index because the electro-optic coefficient involved \( (r_{33}) \) is about three times as large as for the ordinary case \( (r_{13}) \). The change in the ordinary index Fig. 4.2(f) will increase with increased exposure and tend toward the spatial variation already present in Fig. 4.2(e). When crystals were illuminated with a thin strip of light parallel to the c-axis from one c-face to the other, no spatial change in the fringes was observed. The light intensity, in this case, would vary spatially, perpendicular to the c-axis, and be essentially uniform along the c-axis.

4.4 Discussion

The fringe changes, produced by the photorefractive effect, were observed in a Fe-doped (0.015 mole %) crystal, 2.5mm thick. Similar effects were seen in other Fe-doped crystals 1.5mm thick and 10mm thick, however no changes could be seen in the fringe patterns of an undoped crystal (3mm thick). This indicates that the fields induced in undoped crystals are much smaller than those induced in doped crystals.
An estimate of the magnitude of the space charge fields in doped and undoped crystals can be obtained from these photos. To move a dark fringe to the position of the next dark fringe, the optical thickness must change by \( \lambda/2 \). If the thickness, \( d \), remains constant then
\[
\Delta nd = \lambda/2 \quad \text{and} \quad \Delta n = \lambda/2d.
\]
For \( \lambda = 500 \text{ nm} \) and \( d = 2.5 \text{ mm} \), \( \Delta n = 10^{-4} \).

In doped crystals then, \( \Delta n > 10^{-4} \) and in undoped crystals, \( \Delta n < 10^{-4} \).

The space charge field to give \( \Delta n = 10^{-4} \) is
\[
E_{sc} = - \frac{2\Delta n}{r_{33}n_e} = 6.0 \text{ kV/cm}
\]
where \( n_e = 2.24 \) and \( r_{33} = 30.8 \times 10^{-10} \text{ cmV}^{-1} \).

The absence of photorefractive processes when the crystal was illuminated with a strip of light parallel to the c-axis can be attributed to the nature of the electro-optic tensor as was discussed in Chapter 2. The same situation arises when hologram storage is attempted with the plane of incidence of the reference and object beams perpendicular to the c-axis. In both cases the light intensity varies normal to the c-axis and is essentially constant along the c-axis. In our crystals, the optical face contained the a-axis and the c-axis, and the light was transmitted along the b-axis. Space charge fields created along the a-axis would cause only a small rotation of the optical indicatrix and the resulting change in index would be very small.
5.1 Elementary Equations

To probe the mechanism of hologram storage in LiNbO$_3$, the most elementary holographic pattern, the sinusoidal grating, was chosen. This prototype hologram configuration is quite general since any arbitrary intensity variation can be separated into sinusoidal patterns through Fourier decomposition. This type of hologram can be formed when two coherent plane waves interfere within the volume of the crystal. One of these waves is commonly called the reference wave and may be represented by

$$\bar{R} = \text{Re}\left\{ r \exp(i\phi_r) \exp(i\omega t) \right\}$$  \hspace{1cm} (5.1)

The other wave which is commonly called the signal or object wave may be represented in a similar manner by

$$\bar{S} = \text{Re}\left\{ s \exp(i\phi_s) \exp(i\omega t) \right\}$$  \hspace{1cm} (5.2)

where $\omega$ has a single value which is equal for both waves, and $\text{Re}\{\}$ indicates the real part of the complex quantity within the brackets. For convenience, Eq. 5.1 and 5.2 are usually divided by $\exp(i\omega t)$ and the $\text{Re}\{\}$ symbol dropped leaving

$$\bar{R} = r \exp(i\phi_r)$$

and

$$\bar{S} = s \exp(i\phi_s) .$$  \hspace{1cm} (5.3)

The intensity $I$, in the region of interference, is found by taking the scalar product of the sum of the two amplitude vectors

$$I = (\bar{R} + \bar{S}).(\bar{R} + \bar{S})^*$$

$$= r.r + s.s + r.s \exp(i(\phi_s - \phi_r)) + \exp{-i(\phi_s - \phi_r)}$$
or

\[ I = R + S + 2r_s \cos(\phi_s - \phi_r) \]  

(5.4)

where \( R \) and \( S \) are the intensities of the individual waves. The third term in Eq. 5.4 is the interference term which contains the relative phase information. Fig. 5.1 shows the interference of two plane waves. For an angle 2\( \theta \) between the wave normals, the period, \( \ell \), of the sinusoidal intensity distribution is given by

\[ 2 \ell \sin \theta = \lambda \]  

(5.5)

where \( \lambda \) is the wavelength of the light in the medium in which the light is propagating.

Fig. 5.1 Interference pattern of two plane waves.

To write a hologram in LiNbO\(_3\), the crystal is placed in the region of the interference pattern. The spatial variation of the light intensity induces a spatial variation in the refractive indices of the crystal which constitutes a volume hologram. To read out the information stored in the hologram, the crystal is illuminated with a plane wave (the reference wave) and the volume diffraction grating scatters the light in a manner that reconstructs the object or signal wave used
to form the grating. This is shown in Fig. 5.2.

Fig. 5.2 Diffraction of the reference wave by the hologram grating.

Maximum diffraction occurs when Bragg's law (Eq. 5.5) is satisfied.

The diffraction efficiency \( \eta \), of the hologram is the ratio of the diffracted intensity to the incident intensity. Kogelnik (1969) has shown that for thick phase holograms (see Appendix A)

\[
\eta = \exp\left(-\frac{ad}{\cos\theta}\right) \sin^2(\nu d)
\]  

(5.6)

where \( a \) is the absorption, \( d \) is the grating thickness, and

\( \nu = \pi \Delta n/\lambda_o \cos\theta \) for perpendicular polarization and \( \nu = \pi \Delta n \cos2\theta/\lambda_o \cos\theta \)

for parallel polarization. Here, \( \lambda_o \) is the vacuum wavelength and \( \Delta n \) is the amplitude of the sinusoidal refractive index grating. Eq. 5.6 neglects multiple internal reflections between the faces of the crystal. These effects are discussed in the next chapter.

5.2 The Optical System

Fig. 5.3 shows a schematic of the experimental setup used. Light from the laser (either with a Coherent Radiation Model 54 argon ion, or an RCA Model LD2186 He-Cd) was split with a beam splitter into two beams. The transmittance of the beam splitter was variable which
allowed adjustment of the relative beam powers. Two first surface mirrors were used to direct the two beams to cause them to intersect within the volume of the crystal.

The geometry of the setup was such that the path lengths of the two beams from the beam splitter to the crystal were within 1.0 cm of being equal. This ensured that the path difference was less than the coherence length of the laser (10 cm). A silicon photovoltaic detector (Alphametrics model dc 1010 with a PI110 broadband probe) was placed after the crystal in line with the object beam. When the object beam shutter was closed, the energy diffracted from the reference beam toward the detector could be measured, thus allowing the diffraction efficiency of the hologram to be determined.

Fig. 5.4 shows another method used to monitor the diffraction efficiency. An ancillary He-Ne laser was positioned so that the angle of incidence of the beam satisfied the Bragg condition of the phase grating produced by the high power argon ion or He-Cd lasers. As the hologram developed, more and more energy would be diffracted from the incident path of the He-Ne beam allowing continuous measurement of the diffraction efficiency. A low power (2 mw) He-Ne laser (Spectra Physics model 132) was chosen because the photorefractive effect is inefficient with light of wavelength 632.8 nm as compared with light of wavelengths less than 520 nm.

Each of the above methods has its drawbacks experimentally. With the first method, the hologram formation must be interrupted to read the diffraction efficiency. During reading, the reference beam will cause some optical erasure of the hologram. For certain measurements, the optical erasure on readout was reduced by attaching a par-
Fig. 5.3 Experimental arrangement for measuring the diffraction efficiency of plane wave holograms by intermittently blocking the $S$ beam and measuring the intensity diffracted from the $R$ beam.

Fig. 5.4 Alternative arrangement for measuring the diffraction efficiency by continuously monitoring the auxiliary He-Ne beam.
tially silvered glass to an electromagnetic shutter. When the object beam shutter was closed, to facilitate a measurement, the reference beam was attenuated by a factor of approximately 100.

The second method does not erase the hologram or necessitate interruption of its formation but it does present other problems. If the angle of incidence of the He-Ne beam is not very close to the Bragg angle, then the diffraction efficiency of the hologram determined with this beam is much reduced from its true value. Not only is the alignment very critical but determination of the accuracy of the alignment is not an easy task. The first method does not have this alignment problem since the reading and writing beams are the same.

Mechanical stability during hologram formation is another critical experimental consideration. The highest spatial frequency being recorded determines the vibration that may be tolerated. This is generally of the order of the wavelength of the light used for recording. The recording medium must not move more than a fraction of this distance relative to the fringe pattern being recorded. To keep the recording medium steady is not a problem, but to keep the fringe pattern stable special precautions are necessary.

To keep the fringe pattern steady the optical paths of the reference and object beams must remain constant. This means that mechanical vibrations, acoustical and thermal disturbances must be minimized. To accomplish this, experiments were performed on an optical bench. The optical bench had been constructed by epoxying steel strips to a massive concrete base (2.13 x 1.72 x 0.15 m). The table was supported by two columns of cement blocks. Layers of felt were used between each row of blocks to reduce the effects of building vibrations.
To reduce thermal and acoustical disturbance, a plexiglass cover was used to enclose the components on the table top. The laser was left outside the cover because of the heat it generated during operation.

To check the effects of these precautions, a simple interferometer was set up as shown in Fig. 5.5. The detector was masked so that it was illuminated by a portion of one bright interference fringe.

Fig. 5.5 TOP: Arrangement of the interferometer used to check stability. BOTTOM: Variation in the detector signal with time. The line with the arrows indicates 20% of the excursion of the signal when the fringes were made to move past the detector aperture.
5.3 Hologram Storage

With this setup, holograms were easily formed in LiNbO$_3$. Fig. 5.6 shows the build-up of the effective diffraction efficiency (ratio of diffracted intensity to incident intensity), over a period of 4 minutes, of a hologram in a Fe-doped (0.015 mole %) crystal. The saturation effect is typical of all holograms found in this material. This appears to be caused by the space charge field reaching a value where it opposes the process which forms the hologram (Alphonse et al. 1975, Moharam et al. 1975, Gaylord pers. comm.). No attempt was made to estimate the reduced diffraction efficiency due to unstable fringe problems.

![Graph showing the build-up of the effective diffraction efficiency over time](image-url)

Fig. 5.6 Build-up of the effective diffraction efficiency $\eta$, (ratio of incident to diffracted beam power) with time in an Fe-doped (0.015 mole %) crystal.
<table>
<thead>
<tr>
<th>Source</th>
<th>$\eta^2$</th>
<th>$W_0$ (J/cm$^2$)</th>
<th>$a_d$ (cm$^2$/J)</th>
<th>$S$ (cm$^2$/J)</th>
<th>Exposure to get 1% $\eta$ (mJ/cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass et al. 1975b</td>
<td></td>
<td></td>
<td></td>
<td>0.3</td>
<td>330</td>
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<tr>
<td>Alphonse et al. 1975</td>
<td>0.1</td>
<td>0.225</td>
<td></td>
<td>0.444</td>
<td>225</td>
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<tr>
<td>Staebler et al. 1974a</td>
<td>0.122</td>
<td>0.1</td>
<td>1.224</td>
<td>1.22</td>
<td>81.6</td>
</tr>
</tbody>
</table>

(a)

<p>| | | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Undoped Crystal</td>
<td>0.0169</td>
<td>9.94</td>
<td>0.0462</td>
<td>0.367</td>
<td>2700</td>
</tr>
<tr>
<td>Fe doped (0.015M%)</td>
<td>0.059</td>
<td>9.94</td>
<td>0.083</td>
<td>0.715</td>
<td>1400</td>
</tr>
<tr>
<td>Undoped (i)</td>
<td>0.0448</td>
<td>5.676</td>
<td>0.023</td>
<td>0.343</td>
<td>292</td>
</tr>
<tr>
<td>Treated (ii)</td>
<td>0.029</td>
<td>0.8819</td>
<td>0.023</td>
<td>1.43</td>
<td>69.9</td>
</tr>
<tr>
<td>in Li$_2$CO$_3$</td>
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<td>0.251</td>
<td>0.023</td>
<td>2.58</td>
<td>38.7</td>
</tr>
<tr>
<td>(iv)</td>
<td>0.079</td>
<td>0.251</td>
<td>0.023</td>
<td>13.68</td>
<td>7.3</td>
</tr>
</tbody>
</table>

(b)

Table 5.1 Sensitivity of LiNbO$_3$ to hologram storage.

(a) Miscellaneous published data; (b) data obtained from this study.

* (i) Only the central part of the crystal was illuminated, with 0.0 V/cm applied.
(ii) Non-uniform illumination of whole crystal, with 0.0 V/cm applied.
(iii) Crystal nearly uniformly illuminated with 0.0 V/cm applied.
(iv) Crystal nearly uniformly illuminated with 3 kV/cm applied.

Measurements (i) to (iv) are described in Chapter 8.
In Table 5.1 (a), the photorefractive sensitivities of LiNbO$_3$ for various published data are listed. Measurements made during the present study are given in Table 5.1 (b). The change in refractive index $\Delta n$ during hologram recording is linear in the incident energy density $W_0 = \int I \, dt$, in the initial stages. The sensitivity $S$ for small values of diffraction efficiency $\eta$ is

$$S = \frac{\eta}{W_0 \alpha d}$$

(5.7)

where $\alpha$ is the absorption and $d$ the crystal thickness. The ($\alpha d$) factor normalizes the sensitivity for varying amounts of optical absorption in the crystals. Tables 5.1 and 5.2 also list the exposure necessary to achieve 1% diffraction efficiency. There is a wide variation among the different measurements. Some of the variation may be because multiple internal reflections were neglected in the calculations of the diffraction efficiency (see Chapter 6). Other discrepancies may be introduced if the uniformity of the illumination varied among the measurements (see Chapter 8). The most sensitive measurement ($\eta = 1\%$ obtained with 7.3 mJ/cm$^2$) was achieved in an "undoped" crystal with 3 kV/cm applied across the c-faces. Presumably greater sensitivity could be attained by increasing the concentration of Fe$^{2+}$ ions and by applying a larger field.
CHAPTER 6

INFLUENCE OF MULTIPLE INTERNAL REFLECTIONS AND THERMAL EXPANSION ON THE EFFECTIVE DIFFRACTION EFFICIENCY OF HOLOGRAMS IN LiNbO₃

6.1 Introduction

A central question both for engineering applications and for studying the mechanisms of hologram formation is how much diffraction efficiency is produced in given crystals by exposure to known amounts of light. It is shown in this chapter that it is essential to take multiple internal reflections between the crystal surfaces into account when measuring diffraction efficiency. The intensity of the light that is diffracted by the hologram grating is very sensitive to the optical thickness of the crystal. Small changes in temperature such as those produced by the laser beams used in writing the hologram, or fluctuations in the ambient temperature will cause significant changes in the optical thickness. The effects of multiple reflections during the writing process, although probably important, are not considered. Analysis of the effects for illumination with a single beam was given in Chapter 3, but the problem is more complicated for two-beam interaction.

6.2 Theory

The calculations follow the method developed by Kogelnik (1967) in connection with some problems involving the diffraction of light by ultrasonic waves. For simplicity, the analysis is restricted to the case in which the planes of constant refractive index, which constitute the hologram grating, are normal to the optical face of the crystal.
Kogelnik's (1969) coupled wave analysis of diffraction in thick holograms shows that for a beam of unit power density, polarized in the plane of incidence, incident at the Bragg angle $\theta_1$, the energy exchange between the coupled waves leads to a diffracted wave of amplitude

$$S(d) = \frac{\xi}{\cos \theta_1 (\gamma_1 - \gamma_2)} \{\exp(\gamma_2 d) - \exp(\gamma_1 d)\} \exp(-i\beta d) \quad (6.1)$$

where

$$\gamma_1, \gamma_2 = -(\alpha \pm i\xi)/\cos \theta_1$$

$$\xi = -(\pi n_1/\lambda) \cos 2(\theta_1 - \pi)$$

$$\beta = (2\pi n_e/\lambda) \cos \theta_1$$

The crystal thickness is given by $d$, the refractive index by $n_e$, the amplitude of the index modulation forming the grating by $n_1$, and the absorption of the crystal by $\alpha$. This equation assumes that the grating is surrounded by a medium with the same average refractive index.

For gratings formed in crystals, reflections will occur at the boundaries of the grating due to the change in the index at the crystal surfaces. Fig. 6.1 shows the interaction of the multiply-reflected wave with the hologram grating. At the first boundary, the Fresnel transmission coefficient $t_1$ gives the amplitude of the primary wave entering the crystal. After one traversal of the crystal, part of the refracted wave is reflected and a wavelet of amplitude $t_1 t_2 S(d)$ emerges. Both the primary wave and the refracted wave are reflected back into the grating and the interaction continues. The next refracted wavelet to emerge has traversed the grating three times in total and has an amplitude $t_1 t_2 r^2 S(3d)$. Here $t_2$ is the Fresnel transmission coefficient at the second boundary, and $r$ is the Fresnel reflection coefficient for beams incident on the surface from within the crystal. The total
Fig. 6.1 Multiple reflections in a hologram grating.

of all the diffracted wavelets emerging from the crystal is

\[ S_{\text{tot}} = t_1 t_2 S(d) + t^2 S(3d) + t^4 S(5d) + \ldots \] (6.2)

Since \( t_2 = 1 + r \), and \( t_1 = 1 - r \), Eq. 6.2 can be written

\[ S_{\text{tot}} = (1 - R) (S(d) + R S(3d) + R^2 S(5d) + \ldots) \] (6.3)

where \( R = r^2 = \frac{\tan^2(\theta - \theta_1)}{\tan^2(\theta + \theta_1)} \).

Here \( \theta \) is the angle at which the beam is incident on the crystal surface.

If Eq. 6.1 is inserted in Eq. 6.3, two geometric series are obtained

which can be summed to give

\[ S_{\text{tot}} = \frac{S(d)(1 - R) \left[ 1 + R \exp\{d(y_2 + y_1 - 2i\beta)\} \right]}{1 - R \left[ \exp\{2d(y_2 - i\beta)\} + \exp\{2d(y_1 - i\beta)\} \right] + R^2 \exp\{2d(y_2 + y_1 - 2i\beta)\}} \] (6.4)
Fig. 6.2 Effective diffraction efficiency of a hologram in a lithium niobate crystal is plotted against temperature change. For both curves, $n_o(T_c) = 2.252$, $\lambda = 488$ nm, $d = 3$ mm. The solid curve is calculated for no absorption. The dashed line is calculated with $\alpha d = 0.28$. 
Fig. 6.3 Experimentally obtained reflected intensity of an argon ion laser beam incident on a 3 mm thick crystal of undoped LiNbO$_3$ is plotted against time of exposure. The angle of incidence was $15^\circ$ and the total beam intensity was 2360 W/m$^2$. 
Fig. 6.4 Effective diffraction efficiency of a hologram in Fe-doped LiNbO$_3$ is plotted against temperature. The "crosses" are experimentally obtained points. The solid line is calculated with $n_e(T_0) = 2.27351$, $d(T_0) = 1.40$ mm, $\lambda = 441.6$ nm, $ad = 0.252$, $T_o = 31.55^\circ$C, and SS* = 0.5.
The diffracted wave due to multiple reflections is given by

$$S_{\text{tot}} = T S(d)$$  \hspace{2cm} (6.5)

where \(S(d)\) is the wave diffracted in the absence of multiple reflections. The effective diffraction efficiency, \(\eta_e\), defined as the ratio of the intensities of the incident beam external to the crystal and the diffracted beam after leaving the crystal, is

$$\eta_e = SS^* TT^*$$  \hspace{2cm} (6.6)

With no multiple reflections, the diffraction efficiency would just be \(SS^*\).

Since the transmittance factor \(TT^*\) is a function of the path length, the diffracted Fourier components of a real hologram would be differently affected by multiple reflections. In the present case however, the effect of a change in path length can be easily computed. The effect of a change in temperature \((T - T_o)\) can be accounted for by writing the extraordinary index \(n_e\) and the thickness \(d\) as

$$n_e = n_e(T_o)(1 + \delta(T - T_o))$$  \hspace{2cm} (6.7)

and

$$d = d(T_o)(1 + \alpha'(T - T_o)).$$  \hspace{2cm} (6.8)

Reported values of the thermal coefficients are \(\delta = 0.392 \times 10^{-4}/^\circ\text{C}\) (Boyd et al. 1967) at 450 nm and \(\alpha' = 16.7 \times 10^{-6}/^\circ\text{C}\) (Nassau et al. 1966). Hobden and Warner (1966) also give a value for the temperature dependence of the refractive index.

The solid line in Fig. 6.2 shows the variation in the effective diffraction efficiency with changes in temperature for a lossless dielectric grating 3mm thick. The dotted line is for an absorbing grating of the same thickness. For a thicker grating the period of oscillation would be smaller. Very small uniform absorption does not
affect the period of the oscillations, but the maximum effective diffraction efficiency and the amplitude of the oscillations are smaller for the same phase grating. These results were calculated for a refractive index grating that would give a diffraction efficiency of 100% in the absence of multiple reflections and absorption.

In some circumstances, multiple reflections may actually enhance the diffraction efficiency. Fig. 6.3 indicates that a grating that diffracts 10% of the incident light with no reflections can diffract 15% of the light for certain optical paths.

6.3 Experimental Results

Two experiments were performed to determine the effects of temperature on the diffraction efficiency of a simple grating in a 3mm thick, undoped crystal of LiNbO$_3$.

The first experiment was to determine if the beams used to read and write holograms could heat the crystal sufficiently to affect the diffraction efficiency. Fig. 6.4 shows the reflected intensity vs time for an argon ion laser ($\lambda = 488$nm) incident at 15°. The beam was polarized with its electric vector in the plane of incidence and its intensity was 2360 W/m$^2$.

The intensity reflectance of a two-surfaced system is given by (Heavens 1955)

$$R = \frac{r_1^2 + 2r_1r_2 \cos (2\delta_1) + r_2^2}{1 + 2r_1r_2 \cos (2\delta_1) + r_1^2r_2^2}$$

(6.9)

where $r_1$ and $r_2$ are the ordinary Fresnel coefficients for the first and second surface respectively, ($r_2 = -r_1$ for this case) and $\delta_1 = 2\pi n_e d \cos \theta_1 / \lambda$. 
Fig. 6.4 indicates a change in the optical path length $\delta_1$ due to heating. The rate of rise of temperature is initially fast, slowing down as a steady state is approached. A change in temperature will affect $r_1$ and $r_2$ through Eq. 6.7, however the effect is negligible compared to the effect on $\delta_1$ (i.e. the light must be coherent to suffer an intensity change due to temperature fluctuations).

A second experiment was also carried out to show that small changes in the ambient temperature will change the effective diffraction efficiency due to multiple reflections. A hologram was formed in an Fe-doped (0.015 mole%) crystal of LiNbO$_3$ using two plane waves originating from a RCA He-Cd 15 mW laser ($\lambda = 441.6$ nm). The ratio of the reference to the object beam intensity was 1.2 with an angle of incidence between beams of 27.5°. Both beams were polarized with the electric vector parallel to the plane of incidence which also contained the c-axis of the crystal. The crystal was placed in a chamber with the temperature controlled to better than ±0.02°C. The hologram was formed at 35.64 ±0.02°C until the effective diffraction efficiency was about 25%.

After writing the hologram, the effective diffraction efficiency was measured by momentarily exposing the crystal to the reference beam (at reduced intensity) from time to time. Fig. 6.5 indicates that the hologram decayed significantly (due to thermal release of electrons from traps) in the first thirty minutes, after which the thermal decay was small enough not to affect the measurements of interest.

After this initial period, the diffracted light intensity was monitored as the temperature was allowed to slowly fall, and the results are shown in Fig. 6.6. The points are experimental and the solid line was fitted by varying $\eta_e$ and SS* (the absolute diffraction efficiency in Eq. 6.6), with the temperature dependence of $\eta_e$ and d (from Eqs.
The effective diffraction efficiency is clearly sensitive to changes in temperature as small as 1°C. The effect is almost entirely due to a change in the optical path length through the crystal. The angle of refraction only changed about $10^{-3}$ degrees. This would have negligible effect on the Bragg condition. Thermal expansion in the c-direction is eight times smaller than it is in the a or b directions (Nassau et al. 1966) and is, in any case, so small as to cause negligible change in the grating spacing.

In conclusion, it has been shown that a small change in temperature within the LiNbO$_3$ crystal can significantly change the effective diffraction efficiency of a hologram stored in the crystal. A temperature increase can be caused by the absorption of moderately intense laser beams used to read and write holograms. The effect is due to thermal expansion increasing the optical path length of the crystal. Multiple internal reflections cause the diffracted intensity to oscillate as the optical thickness changes.
CHAPTER 7
PHOTOCURRENTS IN LITHIUM NIOBATE

7.1 Introduction

Photocurrents in LiNbO₃ may be measured in the absence of an externally applied field (Chen 1969). As was outlined in Sec. 2.6, Glass et al. (1974b, 1975a) have used the bulk photovoltaic effect to explain this. In this chapter, experimental observations of the photocurrent are given which support the theory that photocurrents will flow in the absence of built-in fields in the crystal.

The experiments to be described show that the relation between the photocurrent and the radiant intensity is linear over the range considered. Cooling crystals from a temperature that relaxes space charge fields, with and without a short applied to the c-faces of the crystal, did not affect the photocurrent. Photocurrents were measured during hologram formation in doped and undoped crystals. An attempt was made to correlate the photocurrent with the induced change in the refractive index.

7.2 Experimental Procedure

For single beam measurements, the crystals were illuminated with an argon ion laser (\( \lambda = 514.5 \) nm). The beam was expanded to illuminate the whole crystal. To make electrical connection to the crystals, gold electrodes were evaporated in the c-faces of the crystals over a flash of chromium. The photocurrent was measured with a Keithley 602 electrometer.

In the holographic measurements, the crystals were illuminated...
in the central portion of the crystal with a beam diameter of 3 mm. The angle between the beams ($2^\theta$) was $30^\circ$. The electric vector of each beam and the c-axis of the crystal were in the plane of incidence. The photocurrent was measured in the same way as the single beam experiments. Both an argon ion laser ($\lambda = 480$ nm) and a He-Cd laser ($\lambda = 441.6$ nm) were used to write holograms.

7.3 Results

When the crystals were illuminated, both a pyroelectric current and a photocurrent were measured. Fig. 7.1 shows the short circuit current measurement on an undoped LiNbO$_3$ crystal. The initial peak is the pyroelectric contribution to the current caused by beam heating. The steady state current is the photocurrent. The photocurrent exhibited no decay after 43 hours of continuous illumination. When the light was turned off, a pyroelectric current of opposite polarity was measured as the crystal cooled. Fig. 7.2 shows the photocurrent for different intensities measured on an undoped crystal. The relationship is clearly linear.

To test the effects of pyroelectric fields on the photocurrent an Fe-doped (0.015 mole %) LiNbO$_3$ was slowly cooled from $375^\circ$C with (i) the c-faces shorted and (ii) with the c-faces open-circuited. In each case the photocurrent was measured after the crystal reached room temperature. The results of three experiments are given in Table 7.1.

Fig. 7.3 shows that in the initial stages of hologram formation, $\arcsin(\eta^2)$ was linear in exposure for an undoped crystal. Fig. 7.4 shows the same result for an iron-doped crystal. From
Fig. 7.1 Time development of the pyroelectric and photo currents during illumination and of the pyroelectric current after the light is turned off.

Fig. 7.2 The photocurrent in an undoped LiNbO₃ crystal for different intensities.
Fig. 7.3 Initial stage of hologram formation in an undoped crystal at two wavelengths. Arcsin(√n) is proportional to the change in index.

Fig. 7.4 Initial stage of hologram formation in an Fe-doped (0.015 mole %) crystal at two wavelengths.
Kogelnik's (1969) derivation of the diffraction efficiency for a sinusoidal grating, a linear relationship is expected since the amplitude of the index modulation is proportional to \( \arcsin \left( \sqrt{\frac{i}{r}} \right) \). The iron-doped crystal produced a greater change in the index than the undoped crystal for the same exposure. As shown in Figs. 7.3 and 7.4, the process was more efficient at \( \lambda = 441.6 \) nm.

<table>
<thead>
<tr>
<th>SEQUENCE</th>
<th>PHOTOCURRENT (pA)</th>
<th>ABSORPTION COEFFICIENT (-1) cm</th>
<th>( I/\alpha ) (pA cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>sc</td>
<td>5.4</td>
<td>0.48</td>
<td>11.3</td>
</tr>
<tr>
<td>oc</td>
<td>3.6</td>
<td>0.31</td>
<td>11.2</td>
</tr>
<tr>
<td>sc</td>
<td>5.8</td>
<td>0.51</td>
<td>11.4</td>
</tr>
</tbody>
</table>

Table 7.1 Effects of short-circuit (sc) and open-circuit (oc) cooling on the photocurrent (\( \lambda = 514.5 \) nm). The absorption was measured on a Cary Spectrophotometer with incoherent light with corrections made for reflections. The crystals were cooled from 375°C to 25°C.

During these measurements the photocurrent was measured after the pyroelectric transient. The currents are listed in Table 7.2.

<table>
<thead>
<tr>
<th>CRYSTAL</th>
<th>( \lambda = 488 ) nm</th>
<th>( \lambda = 441.6 ) nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Doped</td>
<td>0.224</td>
<td>0.216</td>
</tr>
<tr>
<td>Undoped</td>
<td>1.44</td>
<td>0.842</td>
</tr>
</tbody>
</table>

Table 7.2 Photocurrents measured during hologram formation in an Fe-doped (0.015 mole %) and undoped crystal at two different wavelengths.
7.4 Discussion

The measurement of the photocurrent in LiNbO$_3$ shows that it does not decay with time when the entire crystal is illuminated and that it is proportional to the intensity of the illumination. Glass et al. (1974b) have represented the photocurrent density as

\[ J = \kappa \alpha I \]  

(7.1)

where $I$ is the intensity, $\alpha$ the absorption and $\kappa$ a constant depending on the impurities in the crystal. (see Sec. 2.6). From the slope of Fig. 7.2 and with $\alpha = 0.115$ cm$^{-1}$, $\kappa = 1.14 \times 10^{-9}$ Acm/W for the undoped crystal used. This is comparable to Glass et al.'s value of $\kappa = 3.0 \times 10^{-9}$ Acm/W for a Fe-doped crystal.

Although a new transport mechanism is thought to be involved, it is not immediately obvious that the effects of pyroelectric fields can be neglected. It is well known that if no free charges were externally available, LiNbO$_3$ crystals cooled from ~200°C with no field present would develop very large internal and external fields due to the uncompensated change in remanent polarization (Amodei et al. 1972b). These fields are so large that it seems possible that injection or extraction of electrons should set up space charges within the crystal with the result that large scale fields remain in the crystal even after applying an external short. Shorting the c-faces of the crystal during cooling would prevent the development of these space charges. The data in Table 7.1 shows that the photocurrent was independent of the electrical condition during cooling. This experiment failed to confirm the existence of a built-in field. It suggests that built-in fields of pyroelectric origin developed over the temperature range encountered here may be neglected and that some other mechanism is responsible for the photocurrent.
In attempting to correlate the photocurrent measured during hologram formation with the index modulation required to give the measured diffraction efficiency, only the initial region of linear index change was considered. The holograms were considered to be formed by electrons drifting in a field with the migration length being short compared with the grating spacing. The space charge field is given by Eq. 2.7

\[ E_{sc} = \frac{eLg_{mt}}{\epsilon} \cos kx \]  

(7.2)

where \( L = E_o \mu t \). The change in the refractive index is given by Eq. 2.31

\[ \Delta n = \frac{-\kappa^3}{2} \frac{E_{sc}}{\epsilon} \]  

(7.3)

Using Eq. 5.5 and Eq. 6.6 the effects of multiple reflections may be allowed for. The measured build-up in the diffraction efficiency is

\[ \eta_e = \frac{\pi}{2} \sin^2 \left( \frac{\pi \Delta \eta(t) d \cos \theta}{\lambda_0 \cos \theta} \right) \]  

(7.4)

If \( \arcsin \left( \frac{\eta_e}{\pi T^*} \right)^{\frac{1}{2}} \) is plotted as a function of time, the slope of the curve can be calculated from Eq. 7.2 to Eq. 7.4 to be

\[ \text{Slope} = \frac{\pi d \kappa^3 \cos \theta \chi_{33} e g_o L \cos kx}{2 \epsilon \lambda_0 \cos \theta} \]  

(7.5)

Eq. 7.5 may be solved for the expression \( g_o L \). This may be compared with the value of \( g_o L \) computed from the photocurrent in the following way. For a crystal with electrodes separated by a distance \( \lambda \), the average photocurrent measured at the electrodes is assumed to be given by

\[ I_p = e g_o \sqrt{\frac{L}{\lambda}} \]  

(7.6)
where \( g_V \) is the number of electrons (e) generated per second in the illuminated volume (V) and \( L/\ell \) is, on the average, the fraction of the distance each electron travels between the electrodes. From Eq. 7.5 and Eq. 7.6 independent measurements of \( g_L \) can be made.

Table 7.3 lists the values of \( g_L \) calculated by the two methods for a doped (0.015 mole % iron) crystal and an undoped crystal.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Wavelength (nm)</th>
<th>( g_L \times 10^{-12} \text{ m}^{-2} \text{s}^{-1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>from photocurrent</td>
</tr>
<tr>
<td>Undoped</td>
<td>488</td>
<td>9.62</td>
</tr>
<tr>
<td></td>
<td>441.6</td>
<td>5.63</td>
</tr>
<tr>
<td>Doped</td>
<td>488</td>
<td>1.79</td>
</tr>
<tr>
<td></td>
<td>441.6</td>
<td>1.56</td>
</tr>
</tbody>
</table>

Table 7.3 A comparison of \( g_L \), calculated from the photocurrent, to \( g_L \) calculated from the diffraction efficiency.

A comparison of \( g_L \) for different crystals and different wavelengths cannot be made because the intensity of illumination was not the same for all the measurements. There are a number of possible reasons for the discrepancy between the values of \( g_L \) calculated by the two different methods.

The diffraction efficiency measurement gives an estimate of the magnitude of the index modulation. It does not account for any "dc" change in the index which may be produced. As will be shown
in the next chapter, the geometry of illumination of the crystal affects the rate at which holograms are formed. In addition, the optical and thermal history of the crystal may be important. The inclusion of \( T^* \) in Eq. 7.4 to account for multiple reflections provides only an approximate correction since the optical thickness of the crystal is not known to a fraction of a wavelength. Subsequent to these measurements a method has been developed to circumvent this problem (Moharam, Cornish and Young 1975).

Optical erasure of the hologram during its formation may reduce the rate at which the hologram forms. The optical erasure would contribute to the photocurrent but would decrease the index grating. This would cause a discrepancy between \( g_{OL} \) measured using these two methods.

In conclusion, the photocurrent was larger than would be expected from the hologram measurements. A better correlation might be achieved if some of the problems listed above were taken into account.
CHAPTER 8
THE EFFECTS OF INTERNAL AND APPLIED FIELDS ON HOLOGRAMS STORED IN LiNbO$_3$

8.1 Introduction

The application of an external field during the writing of holograms may be an important method of controlling the process. It is also an attractive means of investigating the relative contributions of diffusion, internal fields and the bulk photovoltaic effect to the photorefractive process. The published data on which processes actually occur are in apparent contradiction. Staebler et al. found that either sign of applied field ($\pm 2$ kV/cm (1972b) and $\pm 10$ kV/cm (1974a)) increased the rate of writing equally. This was taken as indicating diffusion. On the other hand, Ohmori et al. (1974) and Yasojima et al. (1972) found that one sign of field increased and the other reduced the rate of generation of optical damage. This was taken as indicating that drift in a built-in field of pyroelectric origin was occurring, as proposed by Chen (1969) and the magnitude of the field was estimated.

The results of the previous experiments by Staebler et al. on the effect of applied voltage, in which holograms were found to be written by diffusion only, assume a new significance in the light of the new theory, the bulk photovoltaic effect, since although pyroelectric fields might be absent in a particular crystal, it is not obvious how the bulk photovoltaic effect should be inoperative.

The experiments to be described show that the results of applied field experiments depend on the fraction of the crystal illuminated and on the intensity of light. In addition, during repeated cycles of hologram writing and reading, with consequent optical erasure, the
diffraction efficiency of the holograms depends on the voltage applied during the previous cycles as well as that applied during the current writing. The explanation of the results is that large-scale "dc" space charge fields (as well as the spatial "ac" fields which produce the holograms) are built-in by exposure to light. The actual field in the crystal depends, therefore, on the field built-in during the previous and current exposure, as well as on the applied field.

An experimental complication is that applied fields of the magnitudes required in these experiments can appreciably affect the optical thickness of the crystal and hence can affect both reading and writing holograms due to multiple internal reflections.

It is well known that exposure to light can "build in" a large scale "dc" field. Although not explicitly taken into account in previous work on the effect of applied voltage, there is no dispute that such fields are created. They are what is observed in compensator experiments (e.g. the original work of Chen (1969)) and in ellipsometer experiments (Chapter 3).

A first point concerns the spatial extent of the "dc" space charge fields relative to the illumination that produces them. As was discussed earlier (sec. 2.3) Chen (1969) explained the results of his circular-beam experiments with a dipole-type field which extended well outside the illuminated area. Chen referred to electrons released and retrapped "outside the illuminated area". This phrase has been quoted or similar statements made in several papers (Johnston 1970, Yasojima et al. 1972, Peterson et al. 1971, Clarke et al. 1973). Actually, the trapping process itself is believed
to be independent of the light intensity. Few electrons are transported outside the illuminated volume and then only for short distances. This was shown in Chapter 3 by experiments in which the light beam effectively varied in intensity only in one dimension. With this geometry, the field and hence the change in birefringence were essentially confined to the irradiated volume.

The next point concerns the effect of how uniformly the crystal is illuminated. For complete, uniform illumination of the crystal (which is impossible with the two plane wave beams used for hologram production) the field in the crystal would be the applied field plus the "virtual" field. With partial illumination a "dc" space charge field will also be present. Thus, with as near as possible the whole crystal illuminated, it is expected that an applied field equal and opposite to the "virtual" field would allow hologram writing by diffusion only. Fields applied with either polarity about this value should increase the rate of hologram production because of increased transport length. If plots of diffraction efficiency vs. voltage show a minimum which is not zero then diffusion may be significant.

8.2 Experimental Procedures

8.2.1 Sample Preparation and Hologram Measurements

The sample used was nominally pure LiNbO₃ (sample #4 of Appendix D) heated in Li₂CO₃ at 520°C for 40 hours, a treatment due to Phillips and Staebler. The absorption was 0.23 cm⁻¹ at 514.5 nm, Holograms were formed using an argon laser (λ = 514.5 nm) polarized parallel

*The "virtual" field (Eᵥ) is the field due to the bulk photovoltaic effect as discussed in Sec. 2.9.1.
to the c-axis with an angle of incidence of 10°. With these crystals, decay of optical damage and of holograms is negligible in the dark (Chapter 3) apparently due to the destruction of shallow traps but holograms are rapidly erased by illumination with the reference beam. It is possible and convenient, therefore, to make repeated experiments and to acquire information on reproducibility, which is absent in previous work. All measurements of the effective diffraction efficiency were made by writing with two plane waves for a specified exposure and then blocking the signal beam and monitoring the reference beam until readout caused the hologram to decay to zero. The magnitude of the effective diffraction efficiency, $\eta$, (ratio of diffracted intensity to incident intensity) was that achieved immediately after the writing and at the commencement of optical readout (Fig. 8.1).

Fields were applied through aluminum electrodes evaporated on to the c-faces of the crystal. The convention is that the direction of the applied field is positive if directed towards the -c face of the crystal. This is in the same direction as the "virtual" field due to the photovoltaic effect.

8.2.2 Multiple Internal Reflections

In the initial work a serious problem became apparent which is not mentioned in earlier work on the effect of applied voltage. This is that both applied voltage and the large scale (as opposed to sinusoidal) space charge fields produced by exposure to light modify the optical thickness of the crystal through the electro-optic effect to a sufficient extent that, because of multiple internal reflections, both the writing and reading efficiency are changed by appreciable amounts. This means that, if this effect is neglected, applied voltage could be interpreted
Fig. 8.1 Measurement of one write, read-erase cycle. The hologram was written for 15 sec with a shutter blocking the detector. A shutter was then used to block the Signal beam and the hologram erasure monitored until the hologram decayed. The effective diffraction efficiency $\eta_e$ given in other figures was that measured at the beginning of read-out i.e. at 15 sec for this example.
Fig. 8.2 Effect of applied field on the transmittance of LiNbO$_3$. The crystal thickness was 1 cm; the angle of incidence of the laser beam was 10° with $\lambda = 514.5$ nm and electric vector parallel to c-axis. Top: crystal after thermal erasure of damage at 270°C. Bottom: the same crystal after hologram writing. The transmittance is diminished because some of the light is diffracted by the hologram. The phase shift along the horizontal axis is due to the optically-induced "dc" space charge field. The bars show the effect of small temperature fluctuations (<0.2°C).
as having an effect on the physics of the electron transport process when in fact it was acting only through the electro-optic effect. The importance of multiple internal reflections was previously pointed out in connection with the measurement of diffraction efficiency (Chapter 6) where they may account for some of the cyclic variations of diffraction efficiency with exposure which have been reported in the literature. The present effect has not previously been mentioned in the literature.

Fig. 8.2 shows the observed transmittance changes due to applied voltages in a crystal 1 cm thick. The upper curve is for a fresh crystal with no optical damage and the lower curve was recorded after a few holograms had been stored in the crystal. The entire curve is lower because some of the incident beam used to record the curve was diffracted by the stored hologram. The main points of interest are the change in transmittance with applied voltage and the phase shift between the two curves caused by the effects of space charge fields induced while writing the holograms. The vertical bars show the fluctuations in transmittance due to small temperature fluctuations (<0.2°C). One solution to the problem of multiple reflections, which was tried initially, is to restrict measurements to voltages giving equal path lengths (modulo λ/2), i.e. equivalent points in Fig. 8.2, for example minima in transmittance. The reading and writing voltages will change with total exposure and must be determined by measuring transmittance or reflectance. An alternative way around the problem of multiple reflections, and the method used in the experiments to be described, is to use sufficiently thin crystals that the change in optical thickness may be neglected. In this case, the effective diffraction efficiency ηₑ still
Fig. 8.3  Effect of prior exposure at different voltages on hologram writing for partial illumination of the sample. The increasing transient shows 3 successive write-read cycles at zero applied field after a 15 sec exposure to the reference beam with $+3 \text{ kVcm}^{-1}$ applied. The decreasing transient shows 3 successive write-read cycles after a 15 sec exposure to the reference beam with $-3 \text{ kVcm}^{-1}$ applied. The bars show the standard deviation of 11 holograms formed at zero applied field. Writing and reading conditions were the same as for the lower curve in Fig. 8.4.
Fig. 8.4 Normalized values of arcsin $\frac{1}{2}$ vs. applied voltage (for electrodes 1 cm apart). The relative area of illumination is shown by the circles and the crystal face (1 cm square) by the squares. Curve A: the exposure corresponded to point 3 on Fig. 8.5 and the intensity was 8.36 mW/cm$^2$. Curve B: the exposure corresponded to point 2 of Fig. 8.5 and the intensity was 44 mW/cm$^2$. Curve C: exposure and intensity, same as B. Curve D: the exposure corresponded to point 1 of Fig. 8.5 and the intensity was 378 mW/cm$^2$. 
differs from the absolute diffraction efficiency, but the difference is
not appreciably changed by the applied voltage or by space charge fields.
It shall be assumed that values of \( \arcsin \sqrt{n_e} \) are a measure of the
relative amplitudes of the refractive index gratings.

8.3 Results

The diffraction efficiency produced by a given exposure was
found to depend on the voltage applied during previous exposures as well as
that applied during the current exposure. This effect was more
pronounced when only a portion of the crystal was illuminated, rather
than the whole crystal. The results shown in Fig. 8.3 were obtained with
two beams of 2.8 mm diameter illuminating the central part of the 1 cm
square face of the crystal. The increasing transient shows the result of
writing at zero voltage (and then reading until the hologram decayed)
three sequential times, after previous illumination with +3 kV/cm applied.
The decreasing transient shows the result of the same experiment but with
-3 kV/cm applied during the previous exposure. The bars represent the
standard deviation of the hologram efficiency of eleven sequential runs
performed at zero applied volts.

Fig. 8.4 shows the effect of applied voltage on the refractive
index grating amplitude per unit exposure, for four different illuminations.
The geometry of illumination for each curve is shown by the circles and
the relative size of the crystal is represented by the squares. Before
each hologram formed at \( \pm \) applied voltage, three or four write, read-
erase cycles were completed at zero applied voltage in an attempt to
establish similar starting conditions and to check for fatigue effects.
Curve A of Fig. 8.4 was measured for exposures shown by point 3 on
Fig. 8.5 and with the beams expanded well beyond the crystal edges so that the intensity was nearly uniform across the crystal. For curve B, the beams were expanded to just illuminate the whole crystal. The exposure for this case corresponded to point 2 on Fig. 8.5. Curve C was measured at the same intensity and exposure as curve B but with only the central portion of the crystal illuminated. This geometry was the same as that for Fig. 8.3. Curve D was measured with the same geometry as curve C but at higher intensity and exposure as indicated by point 11 on Fig. 8.5. The results in Fig. 8.4 show that the efficiency of writing holograms depends on the geometry of illumination and on the intensity. The curves are not symmetrical about zero applied voltage with the asymmetry being more pronounced at higher intensity and with only part of the crystal illuminated.

In one sequence of experiments, over 150 holograms were written in one region of the crystal and read out with no noticeable fatigue in writing efficiency.

8.4 Discussion

In order to illustrate the principal features required to explain the rather complex results described above, a very simple model is considered in which the crystal is uniformly illuminated by a single beam over a length L with dark regions of length \( \xi/2 \) at each side of the illuminated region. Neglecting diffusion, the currents in the illuminated and dark regions, respectively, are:

\[ J(\text{light}) = e\mu nE + n\kappa I \quad (8.1) \]

and

\[ J(\text{dark}) = e\mu n E_a \quad (8.2) \]
Fig. 8.5 The time development of \( \arcsin \eta_e^{1/2} \) during hologram writing for the conditions of curve D of Fig. 8.4. Up to 4 W sec/cm\(^2\) the curves for the reduced intensities (curves A, B and C) in Fig. 8.4 coincided. The numbers indicate the exposures used in Fig. 8.4.
where the free carrier concentration in the illuminated region is 
\[ n = n_0 + g \tau \] and \( n_0 \) is the carrier concentration in the dark. The field in the illuminated region is \( E = E_a + E_{sc} \) and the field in the dark region is \( E_a \) where \( E_a \) is the applied field and \( E_{sc} \) is the space charge field. The discontinuity in the current density at the edges of the illuminated region produces sheets of space charge, \( Q \) per unit area, due to trapped electrons such that

\[
\frac{dQ}{dt} = J(\text{light}) - J(\text{dark}) . \tag{8.3}
\]

The initial value of \( Q \) is \( Q_0 \) and is the charge left from previous exposures. The constraint of constant applied voltage \( V \) gives

\[
(E_a + E_{sc})L + E_a \ell = V \tag{8.4}
\]

where \( \varepsilon E_{sc} = -Q \) and \( \varepsilon \) is the permittivity. Substituting Eq. 8.1 and 8.2 into Eq. 8.3 gives

\[
\frac{dQ}{dt} = \varepsilon \mu (n_0 + g \tau) (E_a + E_{sc}) + \kappa \alpha I - \frac{e\mu n}{\varepsilon} E_a . \tag{8.5}
\]

Substituting for \( E_a \) and \( E_{sc} \) in terms of \( Q \) and \( V \),

\[
\frac{dQ}{dt} = -\left( n_0 + \frac{g \tau L}{\ell + L} \right) \frac{e\mu Q}{\varepsilon} + \frac{e\mu \gamma V}{(\ell + L)} + \kappa \alpha I . \tag{8.6}
\]

The solution of Eq. 8.6 is

\[
Q(t) = Q_\infty \left\{ 1 - \left( 1 - \frac{Q_0}{Q_\infty} \right) \exp\left( -\frac{t}{t_o} \right) \right\} \tag{8.7}
\]

where

\[
t_o = \frac{\varepsilon (\ell + L)}{e\mu \gamma \{ 1 + \frac{n_0 (\ell + L)}{g \tau L} \}} \cdot \tag{8.8}
\]

Substituting the "virtual" field \( E_v \) for \( \kappa \alpha I/e\mu \gamma \), the steady state space
charge field is

\[ E_{sc}(t=\infty) = -\frac{Q_\infty}{\varepsilon} = -\frac{V + E_v(x+L)}{\kappa \left[ 1 + \frac{n_0(x+L)}{g rL} \right]} \quad (8.8) \]

This simplified model shows that the "dc" space charge field adjusts exponentially towards its final value which depends on the applied voltage, the intensity of illumination and the dark conductivity. The field either increases or decreases from the value left from previous exposures.

If a symmetrically placed, smoothly varying light intensity (e.g. Gaussian) is assumed, then, with zero dark conductivity and neglecting diffusion, the steady state space charge field becomes

\[ E_{sc}(x) = -(E_a + E_v)(1 - I(x)/I(0)) \quad (8.9) \]

where \( I(0) \) is the intensity at the electrodes. This steady state space charge field is independent of the intensity of light for a given spatial distribution. If the Glass et al. theory is correct, the dependence found by Chen (1969) on intensity implies that his crystals had appreciable dark conductivity.

The development of the space charge field for a partially illuminated crystal is illustrated in Fig. 8.6, in an idealized way for the first exposure of a crystal (i.e. the initial condition is zero space charge). With the geometry of Fig. 8.6(a), the field (excluding the "virtual" field) is expected to develop as shown in Fig.8.6(b) for an applied field which assists the "virtual" field, and in Fig.8.6(c) for an applied field which just cancels the "virtual" field and in Fig. 8.6(d) for an applied field which opposes and is greater than the "virtual" field. Diffusion has been neglected and the dark conductivity
Fig. 8.6 Idealized illustration of the development of the "dc" space charge field (----) during the first exposure of a crystal. (a) geometry of illumination. (b) for an applied field which assists the "virtual" field. (c) for an applied field which just cancels the "virtual" field. (d) for an applied field which opposes and is greater than the "virtual" field. The applied field is shown by the solid line (——).
has been taken as zero.

For partial or non-uniform illumination of the crystal, if the spatial variation of light intensity could be made the same for erasure and writing, and if the dark conductivity was negligible, then the steady state space charge field left by the erasure process should essentially compensate the "virtual" and applied fields. The more spatially limited the beam, the more exact would be the compensation. The hologram would be written by diffusion, not drift (except for the feedback effect of the sinusoidal space charge field) and applied voltage should have little effect.

With non-negligible dark conductivity the saturation space charge field would depend on the light intensity. These considerations account qualitatively for the diminished effect of an applied field for the experimental conditions of curve D in Fig. 8.4. For curve C of Fig. 8.4 the reduced intensity may have produced only partial compensation of the "virtual" and applied fields by the space charge field, thus showing more dependence on the applied field.

The large scale space charge fields certainly account for the effects of previously applied voltages. When the steady state voltage is changed, the previous steady state space charge field no longer compensates for the applied and "virtual" fields. A few cycles of erasure and writing are required to reach a steady state response, as shown in Fig. 8.3.

From the point of view of applications in read-write memory systems, the above shows that beneficial results would be obtained by suitable changes in applied voltage during writing and erasure cycles.
Curves B and A of Fig. 8.4 show the results of successively more uniform illumination of the crystal. The writing efficiency is greater, the more nearly uniform the illumination because the "dc" space charge field which develops to oppose the writing process is not as large. The voltage which gives minimum efficiency with the crystal as nearly uniformly illuminated as possible (so that space charge fields are minimal) allows the "virtual" field to be estimated, since the minimum should correspond to a cancellation of the applied and "virtual" fields. Values of 0.05 to 1 kV/cm were obtained which are not out of line in view of the wide range (1.5 and 40 kV/cm) reported by Glass et al. (1974b, 1975a) for different crystals. The fact that the minimum efficiency of hologram writing with as nearly as possible uniform illumination was not zero in the above experiments indicates that diffusion may be significant. However, some question exists on this since the spatial patterns of erasure and writing beams are necessarily different and the illumination can be uniform in neither case. It is believed, therefore, that the results are in general agreement with hologram writing due to drift in applied, space charge and "virtual" fields, possibly with some contribution from diffusion.
CHAPTER 9
LUMINESCENCE DUE TO IRON CENTRES

9.1 Introduction

Luminescence in lithium niobate has been observed due to chromium impurities, (Burns et al. 1966, Glass 1969, 1973, Hordvik 1972, 1973), but not, apparently, due to iron, which is the important dopant for the hologram storage applications of this material (Peterson et al. 1973, Amodei et al. 1972). A search was made for luminescence because it was thought that this might help towards understanding the process of hologram writing, which appears to involve a new electron transport mechanism, special to ferroelectrics (Glass et al. 1975b). Absorption of light produces a photocurrent directly, rather than by merely liberating electrons, which then produce current by drifting or diffusing.

9.2 Experimental Procedures

The samples were excited at room temperature using chopped radiation at 325, 488 or 515 nm from a He-Cd or an Argon-ion laser with electric vector along the c-axis. As an indication of the intensity, luminescence was just observable with the naked eye from undoped specimens treated with lithium carbonate. The luminescent radiation was collected with an ellipsoidal mirror and focussed on the entrance slits of a Perkin-Elmer 98G monochromator as shown in Fig. 9.1. Filters were used to prevent laser light entering the monochromator and also, to eliminate incoherent light from the exciting laser beam. The monochromator was fitted with a grating with 300 lines/mm, blazed at 640 nm in
Fig. 9.1 Schematic of the apparatus used to measure the photoluminescence in LiNbO$_3$. 
the first order. The luminescence passing through the monochromator was detected with a cooled S-20 photomultiplier tube and phase-sensitive detector. The spectrometer is interfaced to a NOVA-2 computer for signal averaging and analysis of data. (Thewalt, 1975).

9.3 Results and Discussion

Fig. 9.2 (top) shows the luminescence spectra observed for a congruent crystal doped with 0.015 mole % iron before and after heating the crystal in air to 520°C for 20 h while packed in Li$_2$CO$_3$. A well-defined peak at 770 nm appears following treatment. In Fig. 9.2 (bottom) Li$_2$CO$_3$ treatment of an "undoped" crystal is shown to introduce a small amount of extra luminescence at 770 nm, presumably corresponding to the few ppm of unintentional iron present in this crystal. It is concluded that the peak is due to the presence of iron. It is not understood why the background luminescence was so small in the untreated doped sample. The Li$_2$CO$_3$ treatment reduces iron centres to the Fe$^{2+}$ state. The resultant increase in optical absorption is shown in Fig. 9.3 for the crystals used. A peak or shoulder appears in the region of 470 nm following Li$_2$CO$_3$ treatment. The absorption edge moves to longer wavelengths on adding iron and again following Li$_2$CO$_3$ treatment. The fact that the luminescence peak is so much increased on converting the iron to the Fe$^{2+}$ state suggests that it is due to excitation of these centres. However, the luminescence at 770 nm was larger with the Li$_2$CO$_3$-treated, undoped crystal than with the untreated, doped crystal which showed more absorption at the Fe$^{2+}$ peak at 470 nm, and, therefore, probably contained more Fe$^{2+}$ centres. It has already been mentioned.
Fig. 9.2 Photoluminescence spectra of LiNbO$_3$ at 300$^\circ$K. The excitation wavelength was 325 nm with a power of about 5 mW. TOP: 0.015 mole \% Fe-doped LiNbO$_3$ before and after annealing treatment in Li$_2$CO$_3$. The vertical bars represent the standard deviations for the data after four scans. BOTTOM: Undoped LiNbO$_3$ before and after annealing treatment in Li$_2$CO$_3$. Due to the difficulties in reproducing the optical alignment for different samples, the relative intensities for the four spectra are accurate to only 30 \%. In this and later figures no correction has been made for the S-20 response of the filters.
Fig. 9.3 Absorption spectrum for undoped and for 0.015 mole % Fe-doped LiNbO₃ before and after heating in Li₂CO₃ at 520°C for 20 hours as measured with a Cary spectrophotometer.
that the treatment destroys shallow traps (perhaps due to oxygen vacancies) as well as reducing the iron centres (see Chapter 2). It may be that the luminescence, in fact, involves the trapping of electrons by $\text{Fe}^{3+}$ centres and that the removal of shallow traps means that a higher proportion of excited electrons are captured by the $\text{Fe}^{3+}$ centres.

Fig. 9.4 shows that activation using light of wavelengths typically used for hologram writing also gave the same luminescence peak. In this case, the $\text{Fe}^{2+}$ centre absorption in the region of 470 nm should be involved as opposed to the absorption edge transition (Phillips et al. 1974a, Clark et al. 1973, Redfield et al. 1974). According to Glass et al. (1974b, 1975a) on absorption of light electrons are expelled from $\text{Fe}^{2+}$ centres with momentum in one direction along the c-axis. The question arises as to whether electrons enter the conduction band, or whether they reach an $\text{Fe}^{3+}$ site directly through intervalence transfer. Hush (1967) defines intervalence transfer as "an optical transition which involves transfer of an electron from one nearly localized site to an adjacent one, the donor and acceptor being metal ions which possess more than one accessible oxidation state". In the case where an electron is transferred between a donor and acceptor ion of the same element (homonuclear intervalence transfer) no luminescence is expected (Hush 1967). This, then would not appear to be the case for optical transitions in LiNbO$_3$ involving $\text{Fe}^{2+}$ and $\text{Fe}^{3+}$ ions. Clark et al. (1973) have suggested that intervalence transfer occurs between $\text{Fe}^{2+}$ and $\text{Nb}^{6+}$ ions (heteronuclear intervalence transfer) so that the absorption process is represented as

$$\text{Fe}^{2+}(e) + \text{Nb}^{6+} \rightarrow \text{Fe}^{3+} + \text{Nb}^{5+}(e)$$
Fig. 9.4 Photoluminescent spectra of 0.015 mole % Fe-doped LiNbO₃ at 300°K for two different wavelengths of excitation: 488 nm and 515 nm. The sample was treated in Li₂CO₃. The power of the excitation light was 0.4 W. The relative intensities of the two spectra are accurate to within 10%. Filters were used to block the exciting laser light, and also to block any background luminescence.
They postulate that electrons are then free to move in the conduction band made up of the niobium d orbitals until they are retrapped by Fe\textsuperscript{3+} ions. In this case, the luminescence which was observed would occur when the excited electrons were retrapped.

In conclusion, the observed luminescence due to iron centres would appear to indicate that electrons are distributed by means other than homonuclear intervalence transfer between Fe\textsuperscript{2+} and Fe\textsuperscript{3+} sites. The details of the process which are involved however, are not presently understood. The occurrence of homonuclear intervalence transfer of course is not ruled out, particularly at higher iron concentrations.
The purpose of this work was to study the mechanisms of the photorefractive effect to further the understanding of the process for engineering applications. The course of the work was briefly as follows. Initially, the phenomenon was investigated to clarify the role of drift and diffusion as charge transport mechanisms. Later, when the bulk photovoltaic effect was proposed as a transport mechanism, further experiments were carried out to investigate this effect. In addition, studies were made to extend the usefulness of automated ellipsometry in probing the photorefractive process. The effects of applied fields on the photorefractive effect, and the effects of multiple internal reflections on techniques used to probe this effect, were also investigated.

The contributions which were made to the subject may be summarized as follows:

a) A theoretical treatment of the development of refractive index gratings through drift and diffusion was made without the restriction of short migration length. The principal result was that the efficiency of hologram writing increases for increased migration length up to a certain limit. It was also shown that the increased migration length would not limit the resolution of the recording medium.

b) An explanation was provided for the apparent contradiction which existed in the published data from applied field experiments. It was shown that the results of such experiments depend on the field applied during previous exposure to light as well as on the field applied during the current experiment. In addition, the measurements depended on the
portion of the crystal illuminated since this affected the magnitude of
the space charge field that could be developed. The results were con-
sistent with holograms formed by drift, diffusion and the bulk photo-
voltaic effect. It was found that either sign of the applied field could
increase the efficiency of hologram formation but the effect was not
symmetrical. The asymmetry was attributed to the photovoltaic effect.
No value of the applied field would totally inhibit hologram formation
indicating that diffusion was partially responsible.
c) Further evidence was given for the existence of the bulk photovoltaic
effect. It was shown that the photocurrent in lithium niobate was not
primarily due to pyroelectric fields developed during cooling of the
crystals.
d) The importance of multiple internal reflections on the interpretation
of experimental data was recognized. It was shown that the effective
diffraction efficiency measured outside the crystal can vary significantly
from the absolute diffraction efficiency of the phase grating. Small
changes in temperature such as those produced by exposure to medium inten-
sity laser beams produce sufficient change in crystal thickness to have
a significant effect. Multiple reflections were also shown to be impor-
tant in applied field experiments and in the interpretation of ellipsometry
and adjustable-compensator measurements.
e) The advantages of using an ellipsometer to probe the birefringence
and optically induced changes in the birefringence of lithium niobate
crystals were investigated. This was practicable since the ellipso-
meter was automated through computer control. In addition to using
ellipsometry, it was shown that large scale changes in the refractive
indices could be rapidly inspected if the crystal was made to act as a
Fabry-Perot interferometer.

f) It was shown that when lithium niobate crystals are heated in lithium carbonate, the treatment reduces iron impurities, changes the birefringence of the crystal and decreases the rate at which optically induced space charge fields decay. To explain these results, it was proposed that the treatment destroys shallow traps.

g) Luminescence due to iron centres was observed for the first time. The spectral behaviour of the luminescence was not understood but would be related to the redistribution of optically excited electrons among traps.

10.1 Suggestions for Further Research

Further investigations are required to characterize more completely the mechanisms of the photorefractive effect. Some of the parameters involved in charge transport which are not presently known are the quantum efficiency of photo-excitation, the lifetime of free carriers, the capture cross section of traps, and the migration length of free electrons. These parameters are undoubtedly influenced by the concentration of defects and impurities in the crystal. Additional work is required on the nature of the defects and their control. It was suggested in Chapter 3 that heating lithium niobate in lithium carbonate destroys shallow traps. Identification of these traps and how they influence the photorefractive effect would be useful.

The luminescence studies outlined in this thesis could usefully be extended. Information on the lifetime of excited electrons and the energy levels of the traps involved could possibly be obtained.
In applications where increased photorefractive sensitivity is necessary, research into optimizing the mobility and lifetime of free electrons is required. Possibly, materials other than lithium niobate would have an advantage in this instance.
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APPENDIX A

FURTHER PROPERTIES OF LITHIUM NIOBATE

A.1 Miscellaneous Physical Properties

The structure of the ferroelectric crystal lithium niobate is rhombohedral, point group symmetry 3m with \( a = 0.5499 \text{ nm} \) and \( \alpha = 55^\circ 52' \) (Nassau et al. 1966a). At room temperature the crystalline structure consists of planar sheets of oxygen atoms in approximately hexagonal close packing. The resulting octahedral interstices are one-third occupied by Nb and one-third by Li with the remainder vacant (Abrahams et al. 1966a, 1966b). All the oxygen octahedra are distorted and there are two values each for the Li-O and Nb-O distances. Abrahams et al. conclude that lithium niobate is not primarily an ionic crystal but one in which covalent bonds dominate.

The pure crystal has very little absorption from 350 nm to 5\( \mu \)m. It is a uniaxial crystal with ordinary and extraordinary refractive indices at 500 nm reported to be \( n_o = 2.34 \) and \( n_e = 2.24 \) at 25°C. The dispersion and temperature dependence of the refractive indices has been reported (Boyd et al. 1964, 1967, Miller et al. 1965, Hobden et al. 1966, Nelson et al. 1974). The dielectric constant has been measured for directions perpendicular to the c-axis as 78 and along the c-axis as 32 (Nassau et al. 1966). High temperature transport processes have been measured by Jorgensen and Bartlett (1969) who found that both ionic and electronic conductivity occur. The electrical conductivity is completely ionic at 1 atm of oxygen and 1000°C, while for low oxygen partial pressures the conductivity at 1000°C becomes completely electronic. The electronic conductivity is proportional to \( p_{O_2}^{-k} \).
The electron mobility was calculated to be $1.7 \text{ cm}^2/\text{Vsec}$ at $1000^\circ\text{K}$ in a 50% CO/50% CO$_2$ atm and exhibits a $T^{-2/3}$ temperature dependence. The ionic conductivity is cationic and is most probably due to transport by lithium ions. The Curie temperature is $1210^\circ\text{C}$ and the melting point about $1260^\circ\text{C}$ (Nassau et al. 1966). At room temperature the crystal is a stable ferroelectric. The pyroelectric coefficient is $10^{-2} \mu\text{C/}(\text{m}^2\text{deg})$ in the region of $100^\circ\text{C}$ (Roitberg et al. 1970). The coefficients of thermal expansion are $16.7 \times 10^{-6}$ per $^\circ\text{C}$ in the $a$-axis direction and $2 \times 10^{-6}$ per $^\circ\text{C}$ in the $c$-axis direction.

A.2 Crystal Growth

Nassau et al. (1966b) have described techniques used to grow single crystals of lithium niobate. The most common is the Czochralski technique with an electric field applied during growth (Fig. A.1). The crystal is rotated as it is pulled from the melt. If no field is applied during growth a multi-domain crystal forms. Either polarity may be applied, however if polarity is reversed during growth a $180^\circ$ domain wall is produced. Crystals which are not poled during growth may be poled afterwards, but only at temperatures above $1000^\circ\text{C}$.

A.3 Thermal Bleaching and Fixing of Holograms in LiNbO$_3$

Optical erasure of holograms which usually occurs during read-out can be avoided if the holograms are fixed (Amodei et al. 1972a, Staebler et al. 1972a). When a crystal in which a hologram has been stored is heated to $100^\circ\text{C}$ for 20 or 30 minutes and then cooled, it is found that the hologram has been bleached. The phase grating may be restored by illuminating the crystal with light of wavelengths 400 to 500 nm. The
Fig. A.1 Apparatus for the Czochralski growth of lithium niobate in an electric field (Nassau et al. 1966b).
restored grating cannot be optically bleached.

Amodei et al. have proposed that the hologram is bleached not by thermally activated electrons which are redistributed uniformly, but by some kind of ionic movement which compensates the space charge. When the crystal is cooled and then illuminated, optically excited electrons redistribute uniformly. The phase grating reappears due to the ionic displacements which are insensitive to light.

Qualitatively, this explanation accounts for the experimental results. Further investigation of the mechanisms involved in electronic and ionic transport are required before details of the thermal processes in lithium niobate can be fully understood. If the ionic motion at 100°C is due to lithium ions, it may be that the charge compensation is caused by lithium ions migrating to vacant oxygen octahedral interstices normally present in the crystal (Abrahams et al. 1966a). When the crystal is cooled these ions are frozen in their new sites in the crystal lattice.
APPENDIX B

ELECTRO-OPTIC BEHAVIOUR OF LITHIUM NIOBATE

The propagation of electromagnetic waves in an anisotropic dielectric crystal is dependent on the propagation and polarization direction of the wave with respect to the crystal axes. The dielectric properties at optical frequencies are given by

\[ D_i = \varepsilon_0 \varepsilon_{ij} E_j \]  

(B.1)

where \( D \) is the displacement, \( E \) is the electric field, \( \varepsilon_0 \) the permittivity of free space and \( \varepsilon_{ij} \) the tensor permittivity of the medium. Combining Eq. 1 with Maxwell's equations leads to the conclusion that two waves of different velocities may, in general, propagate through the crystal for a given wave normal (Nye 1960). The refractive indices of the two waves may be obtained by drawing an ellipsoid known as the indicatrix. If \( x_1 \), \( x_2 \) and \( x_3 \) are the principal directions of the permittivity tensor, the indicatrix is defined by the equation

\[ \frac{x_1^2}{n_1^2} + \frac{x_2^2}{n_2^2} + \frac{x_3^2}{n_3^2} = 1 \]  

(B.2)

where \( n_1 = \sqrt{\varepsilon_1} \), \( n_2 = \sqrt{\varepsilon_2} \), \( n_3 = \sqrt{\varepsilon_3} \).

If a straight line is drawn from the centre of the ellipsoid parallel to the wave normal of the propagating wave, then an ellipse may be formed by cleaving the ellipsoid through its centre, perpendicular to this line. The semi-axes of this ellipse define the two directions of polarization which may propagate. The indices of refraction seen by the two propagating waves are then given by the length of the semi-axes.

If an electric field is present, the refractive index of the
crystal is altered and the new indicatrix is in general described by

\[ \sum_{i,j;k,l} \left[ \frac{1}{2} + z_{ijk} E_k + R_{ijkl} E_k E_l + \ldots \right] x_i x_j = 1 \]  

where the indices \( i,j,k,l \) run from 1 to 3. The coefficients \( z_{ijk} \) and \( R_{ijkl} \) are the linear and the quadratic electro-optic coefficients.

Contractions in the indices are usually made as follows: \( r_{mk} \leftrightarrow z_{(ij)k} \) and \( R_{mn} \leftrightarrow R_{(ij)(k\ell)} \) where \( m \) and \( n \) run from 1 to 6 and \( m \) is related to \( (ij) \) and \( n \) to \( (k\ell) \) as follows: 1 \( \to \) 11, 2 \( \to \) 22, 3 \( \to \) 33, 4 \( \to \) 23, 5 \( \to \) 13, 6 \( \to \) 12.

Certain systems cannot exhibit the linear electro-optic effect (such as those with a centre of symmetry) while all materials exhibit the quadratic effect. Lithium niobate exhibits the linear electro-optic effect. Symmetry considerations require that some of the linear electro-optic coefficients are equal and that some are zero as shown by the following matrix (class 3m).

\[
\begin{bmatrix}
0 & -r_{22} & r_{13} \\
0 & r_{22} & r_{13} \\
0 & 0 & r_{33} \\
0 & r_{42} & 0 \\
r_{42} & 0 & 0 \\
-r_{22} & 0 & 0 \\
\end{bmatrix}
\]

where (Turner 1966) \( r_{13} = 8.6 \times 10^{-10} \text{ cm/volt} \), \( r_{22} = 3.4 \times 10^{-10} \text{ cm/volt} \), \( r_{42} = 28 \times 10^{-10} \text{ cm/volt} \), \( r_{33} = 30.8 \times 10^{-10} \text{ cm/volt} \).
A further property of LiNbO$_3$ is that it is a uniaxial crystal with $x_3$ considered as the polar axis. Hence, the indicatrix is an ellipsoid of revolution and two of the three principal semi-axes are equal so that

$$n_0 = n_1 = n_2, \quad n_e = n_3.$$ 

The indicatrix is thus given by

$$\frac{1}{n_o} - r_{22} E_2 + r_{13} E_3 \right) x_1^2 + \frac{1}{n_o} + r_{22} E_2 + r_{13} E_3 \right) x_2^2 
+ \frac{1}{n_e} + r_{33} E_3 \right) x_3^2 + 2(-r_{22} E_1) x_1 x_2 
+ 2(r_{42} E_2) x_2 x_3 + 2(r_{42} E_1) x_3 x_1 = 1. \quad (B.4)$$

From this equation it can be seen that if $E_3$ is the only field present, then only an extension or contraction of the major axes is possible. Since all the cross terms would be zero, no rotation of the principal axes of the indicatrix occur. If however $E_1$ or $E_2$ are present then a rotation occurs.

For a wave propagating in the $x_2$ direction, with only $E_3$ present, the equation of the indicatrix reduces to

$$\frac{1}{n_o} + r_{13} E_3 \right) x_1^2 + \frac{1}{n_e} + r_{33} E_3 \right) x_3^2 = 1 \quad (B.5)$$

The effect of $E_3$ is to introduce changes, $\Delta n_o$ and $\Delta n_e$ in the two refractive indices.

Manipulation of equation (B.5) shows that

$$\Delta n_o = -\frac{n_o^3 r_{13} E_3}{2} \quad \text{and} \quad \Delta n_e = -\frac{n_e^3 r_{33} E_3}{2} \quad (B.6)$$

The change in index is then proportional to the field.
APPENDIX C

COUPLED WAVE THEORY FOR THICK HOLOGRAM GRATINGS

Kogelnik (1969) has treated the Bragg diffraction of plane waves by thick phase holograms using coupled wave theory. A condensed version of his treatment will be outlined for the case of perfect Bragg conditions and for the case where the grating planes are parallel to the bisector of the two incident beams R and S, as indicated in Fig. C.1.

![Diagram](image)

Fig. C.1 Model of a thick hologram grating of thickness d, grating spacing λ and Bragg angle, in the medium, θ.

The analysis assumes monochromatic light to be incident on a phase hologram grating of thickness d at an angle θ polarized perpendicular to the plane of incidence. Only two waves are assumed to be present: the reference wave R and the signal wave S. These two waves are the only waves obeying the Bragg condition. The assumption limits the analysis to thick
holograms. (Storck and Wolff (1975) have shown that, in the case of low
diffraction efficiency, the analysis is also valid for thin holograms).

Wave propagation in the grating is described by

\[ \nabla^2 \vec{E} + k^2 \vec{E} = 0 \]  \hspace{1cm} (C.1)

The propagation constant is related to the relative dielectric constant 
\( \varepsilon \), and the conductivity \( \sigma \), by

\[ k^2 = \frac{\omega^2}{c^2} \varepsilon - i\omega\mu\sigma \]  \hspace{1cm} (C.2)

where \( c \) is the velocity of light, \( \mu \) is the permeability of the medium
and \( \omega \) the angular frequency of the electric field \( \vec{E}(x,z) \). The fringes
of the phase grating result from a spatial modulation of \( \varepsilon \):

\[ \varepsilon = \varepsilon_o + \varepsilon_1 \cos kx \]  \hspace{1cm} (C.3)

where \( \varepsilon_1 \) is the amplitude of the spatial modulation and \( \varepsilon_o \) is the average
dielectric constant. (The spatial modulation in the conductivity is
assumed to be negligible). The grating vector \( k \), in the case under
consideration has only an \( x \)-component and is given by

\[ k = \frac{2\pi}{\lambda} \]  \hspace{1cm} (C.4)

where \( \lambda \) is the grating period and is related to the wavelength of light
\( \lambda \) by \( \lambda = 2\pi \sin \theta \).

Eqs. C.2 and C.3 can be combined to give

\[ k^2 = \beta^2 - 2i\alpha \beta + 2\kappa \beta (e^{ikx} + e^{-ikx}) \]  \hspace{1cm} (C.5)

where \( \beta \) is the average propagation constant and \( \alpha \) the average absorption
constant:

\[ \beta = \frac{2\pi \varepsilon_o^{\frac{1}{2}}}{\lambda}; \quad \alpha = \frac{\mu c \sigma}{2\varepsilon_o^{\frac{1}{2}}} \]  \hspace{1cm} (C.6)
The coupling constant \( \kappa' \) is defined as

\[
\kappa' = \frac{\pi \epsilon_1}{2 \lambda \epsilon_0^{1/2}} \tag{C.7}
\]

The coupling constant describes the coupling between the reference and the signal wave.

Optical media may be characterized by their refractive index \( n \), when the following conditions are met.

\[
\frac{2\pi n}{\lambda} \gg \alpha; \quad n \gg n_1 \tag{C.8}
\]

Here \( n \) is the average refractive index and \( n_1 \) the amplitude of the spatial modulation. These conditions will be assumed to be met, in which case

\[
\beta = \frac{2\pi n}{\lambda}; \quad \kappa' = \frac{\pi n_1}{\lambda} \tag{C.9}
\]

The spatial modulation in the refractive index forms a grating which couples the two waves \( R \) and \( S \) and causes an exchange of energy between them. The complex amplitudes of these two waves, \( R(z) \) and \( S(z) \) vary along \( z \) as a result of the energy interchange and because of absorption. The total electric field in the grating is

\[
E(x,z) = R(z) \exp(-i\vec{\rho} \cdot \vec{x}) + S(z) \exp(-i\vec{\xi} \cdot \vec{x}) \tag{C.10}
\]

where

\[
\vec{x} = \begin{bmatrix} X \\ Y \\ Z \end{bmatrix}; \quad \vec{\rho} = \begin{bmatrix} \sin \theta \\ 0 \\ \cos \theta \end{bmatrix}; \quad \vec{\xi} = \begin{bmatrix} \sin(-\theta) \\ 0 \\ \cos(-\theta) \end{bmatrix}
\]

To solve the coupled wave equations, Eq. C.1, C.5 and C.10 are combined.

By comparing terms with equal exponentials (\( e^{-i\vec{\rho} \cdot \vec{x}} \) and \( e^{-i\vec{\xi} \cdot \vec{x}} \))

*The results of this analysis may be applied to parallel polarization by substituting \( \kappa'' \) for \( \kappa' \) where

\[
\kappa'' = \kappa' \cos 2\theta
\]
we arrive at

\[ R'' - 2iR' \beta \cos \theta - 2i\alpha R + 2\kappa BS = 0 \]  \hspace{1cm} (C.11)

and

\[ S'' - 2iS' \beta \cos \theta - 2i\alpha S + (\beta^2 - \sigma^2)S + 2\kappa BS = 0 \]  \hspace{1cm} (C.12)

where the primes indicate differentiation with respect to \( z \). If the interaction between the \( R \) and \( S \) beams is slow then the \( R'' \) and \( S'' \) terms may be neglected. Eq. C.11 and C.12 can be rewritten in the form

\[ R' \cos \theta + aR = -i\kappa S \]  \hspace{1cm} (C.13)

\[ S' \cos \theta + aS = -i\kappa R \]  \hspace{1cm} (C.14)

Physically, the \( R \) and \( S \) waves change their amplitude along \( z \) due to coupling to the other wave (\( \kappa R, \kappa S \)) and due to absorption (\( aR, aS \)).

The general solution to the coupled wave equations is

\[ R(z) = r_1 \exp(\sigma_1 z) + r_2 \exp(\sigma_2 z) \]  \hspace{1cm} (C.15)

\[ S(z) = s_1 \exp(\sigma_1 z) + s_2 \exp(\sigma_2 z) \]  \hspace{1cm} (C.16)

The constants \( r_1 \) and \( s_1 \) depend on the boundary conditions. The constants \( \sigma_1 \) may be obtained by substituting Eq. C.15 and C.16 into the coupled wave equations. The solution is

\[ \sigma_{1,2} = -\frac{1}{\cos \theta} (\alpha \pm i2\kappa) \]  \hspace{1cm} (C.17)

To find \( r_1 \) and \( s_1 \) the boundary conditions for transmission hologram are introduced. The reference wave \( R \) is assumed to start with unit amplitude at \( z = 0 \). As it propagates through the phase grating it decays as it couples energy into \( S \) which is assumed to be zero at \( z = 0 \). The boundary conditions are

\[ R(0) = 1, \quad S(0) = 0 \]  \hspace{1cm} (C.18)

Solving for \( r_1 \) and \( s_1 \) and substituting in Eq. C.16 the amplitude of the signal wave as it leaves the grating is given by
\[ S(d) = \frac{1}{\cos (\gamma_1 - \gamma_2)} (\exp(\gamma_2 d) - \exp(\gamma_1 d)) \]  
(C.19)

where \( \gamma_{1,2} = -(\alpha \pm i\kappa) / \cos \theta \). The diffraction efficiency of the grating \( \eta \) is defined as

\[ \eta \equiv SS^* . \]  
(C.20)

In the present case Eq. C.20 reduces to

\[ \eta = \exp(-\frac{2\alpha d}{\cos \theta}) \sin^2 \nu d \]  
(C.21)

where \( \nu = \pi n_1 / \lambda \cos \theta \) for perpendicular polarization and \( \nu = \pi n_1 \cos 2\theta / \lambda \cos \theta \) for parallel polarization.
APPENDIX D

SOURCES OF THE LITHIUM NIOBATE CRYSTALS

The crystals used in this study were obtained from Crystal Technology, Inc., Mountain View, California and from Harshaw Chemical Company, Solon Ohio. Table D.1 lists the nominal dimensions, orientation and impurity doping of the crystals. Crystals 1, 2 and 3 were cut from Crystal Technology boule #10-375 which was grown along the c-axis. Crystal 4 was cut from boule #10-286 which was grown along the b-axis. The fifth crystal was purchased from Harshaw.

All the crystals were grown by the Czochralski technique. The composition of the melt from which the crystals were grown is given in Table D.1. A stoichiometric melt contains more Li than does a congruent melt (Redfield et al. 1974). In our crystals, the stoichiometric melt was 49.0 mole% Li$_2$O while the congruent melt was 48.6 mole% Li$_2$O.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Dimensions (mm)</th>
<th>Polished face</th>
<th>Iron-doping (mole %)</th>
<th>Composition of the melt</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>a</td>
<td>b</td>
<td>c</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>a</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>2.5</td>
<td>20</td>
<td>b</td>
</tr>
<tr>
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<td>1.0</td>
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<td>b</td>
</tr>
<tr>
<td>5</td>
<td>15</td>
<td>3</td>
<td>20</td>
<td>b</td>
</tr>
</tbody>
</table>

Table D.1

Lithium niobate crystals used in this study.
APPENDIX E

THE APPLICATION OF LITHIUM NIOBATE

IN A HOLOGRAPHIC MEMORY SYSTEM

Read-write optical memories based on hologram storage in an erasable medium are of interest because they offer the possibility of large storage capacity with little or no mechanical motion and high speed random accessibility. Access to data would be on a page-by-page basis with possibly \(10^5\) bits read or written in parallel. The basic components for a holographic memory are shown in Fig. E.1. An array of light valves composes the page of data to be stored. For writing, the signal and reference beams are directed to the appropriate position on the holographic storage medium by beam deflectors. For reading, the reference beam would be diffracted on to a sensor array by the hologram. Erasure could occur simultaneously during the reading or writing cycles or could be accomplished separately. The page composer and sensor arrays would be electrically addressable by the computer.

Possible devices that could be used to deflect the beam are acousto-optic and electro-optic deflectors. The acousto-optic deflector is the simpler of the two but it is slower. The electro-optic deflector is more complicated and presently requires high operating voltages. Both these devices can be made with LiNbO\(_3\) crystals (Korpel et al. 1966, Chen 1970).

The system is arranged so that all the stored holograms diffract the reference beam on to a single detector array. The detector may be an array of photodiodes produced as an integrated circuit. Economically it may be more feasible to make up the detector array with
Fig. E.1. (a) Schematic of a read, write, erase in-situ holographic optical memory. In this configuration, the page composer is used in reflection. Page composers may also be used in transmission. (b) Schematic of readout in a page-organized holographic memory.
a number of small integrated circuits.

The laser power required is determined by the number of elements in a page and the efficiency of the optical system. For instance, in a system in which the holographic diffraction efficiency was 10% and the transmission efficiency of all other components was 20%, then only 2% of the laser power would reach the detector array. If this was then shared between $10^4$ detector elements only about $10^{-6}\%$ of the laser power would reach each element. For a detector that required 1 pJ of light, a reading speed of 1 μs per hologram would require a laser with a power of 1 W.

The page composer envisioned in this type of system would consist of an array of elements that could be switched between a transparent condition and an opaque condition to correspond to ones and zeros. A number of different types of page composers have been investigated but only prototypes have been built. Some experimental holographic systems have used nematic liquid crystals (Stewart et al. 1973, d'Auria et al. 1974). These require a buffer memory and at least one electrical connection for each bit. In addition, they are inherently slow, typically requiring several milliseconds to switch states. Ferroelectric ceramics such as PLZT have also been considered for use in page composers. Transparent electrode strips are deposited on each face of a slice with parallel strips on the one side being orthogonal to those on the other side. Coincident addressing changes the birefringence between the crossed electrodes. For an $N \times N$ array of elements only $2N$ electrical connections are required. Although the devices are faster than liquid crystals, at present the PLZT fatigues both electrically and optically.
Carlsen (1974) has proposed an alternative method for recording the digital data. In his system data bits would be stored sequentially as they arrived from the computer thus alleviating the need for a large page composer. The advantages of parallel input are lost unless the data to be stored in one page is prearranged. Otherwise the data stored in any one page would be so diverse that there would not be much advantage in retrieving it all in a parallel output mode. With random access sequential input, the computer could tag the data for storage in a given page. In this manner, parallel output of related data could be achieved. Carlsen proposed that each bit in a page be stored with a different angle with all bits in a page superimposed upon one another to yield a multiple exposure hologram. Each page would be stored in a different location in the recording medium. In using multiple exposure holograms, the need may arise to selectively erase some of the superimposed bits but not others. Huignard et al. (1975) have demonstrated that this is possible. Using LiNbO$_3$ as a storage medium, they first stored a number of holograms in superposition. To erase a given hologram they recorded the same object again but with the reference wave shifted in phase by $\pi$. This produced a complimentary spatial modulation in the refractive index thus eliminating that particular hologram. From a practical point of view this method would not be very useful since extreme stability would be required.

It is of interest to compare holographic memory systems with other systems to determine what role they might play. Kiemle (1974) has shown that holographic memories using a single detector array and a single hologram plate are limited in storage capacity to about $10^8$. 

New memory techniques such as charge-coupled devices and magnetic bubble domains may be capable of this capacity range and may provide cheaper solutions. Conventional recording technologies such as magnetic drums and disks will probably be improved. Development of holographic memories therefore should strive to complement these and other technologies because it is unlikely that they will completely replace them. It is envisioned that holographic memories will be able to provide capacities comparable to those of magnetic tape storage systems, but with much shorter access times.

The capacity limit mentioned above, of $10^8$ bits for holographic memories using a single detector array and recording plate, arises because the angle at which the detector array can be illuminated is limited. This determines the maximum size of the recording plate. Kiemle (1974) however has investigated the concept of using modules which each contain a page composer, hologram storage medium and detector. The memory system would have one laser, one x-y beam deflector and a number of passive beam dividers. This concept would allow random access to virtually unlimited capacity. In Table E.1 a comparison is made of some memory systems.
<table>
<thead>
<tr>
<th>MEMORY TYPE</th>
<th>CAPACITY (bits)</th>
<th>RANDOM ACCESS TIME</th>
<th>DENSITY</th>
<th>REFERENCE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Semiconductor</td>
<td>$10^9-10^{10}$</td>
<td>&lt; 1 ms</td>
<td>2000 bits/cm$^3$</td>
<td>Hodges (1975)</td>
</tr>
<tr>
<td>Core</td>
<td>$10^6$</td>
<td>1 μs</td>
<td></td>
<td>Rajchman (1970)</td>
</tr>
<tr>
<td>Disk</td>
<td>$10^8$</td>
<td>100 ms</td>
<td>1.4x$10^5$ bits/cm$^2$</td>
<td>Matick (1972)</td>
</tr>
<tr>
<td>Magnetic Tape</td>
<td>$10^{12-10^{13}}$</td>
<td>&gt; 5 s</td>
<td>2.4x$10^5$ bits/cm$^2$</td>
<td>Wildmann (1975)</td>
</tr>
<tr>
<td>(IBM TBM System)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IBM 3850 System</td>
<td>$10^{12}$</td>
<td>15 s</td>
<td>2.5x$10^6$ bits/cm$^3$</td>
<td>Harris et al.(1975)</td>
</tr>
<tr>
<td>Magnetic Bubbles</td>
<td>$10^6-10^8$</td>
<td>.5-5 ms</td>
<td>16 x$10^6$ bits/cm$^2$</td>
<td>Bobeck et al.(1975)</td>
</tr>
<tr>
<td>Holographic Memories</td>
<td>&gt; $10^{13}$</td>
<td>1 μs-1 ms</td>
<td>10$^8$ bits/cm$^2$</td>
<td>Kiemle (1974)</td>
</tr>
</tbody>
</table>

Table E.1

A comparison of some computer memory systems.
APPENDIX F
ELLIPSOMETER ALIGNMENT

The ellipsometer is a geometrical instrument which depends on the relative alignment of its components for its accuracy. Errors may arise from zero errors in the azimuth scales of the polarizer, quarter wave plate and analyzer, from a zero error in the angle of incidence scale and from imperfections in the optical elements, especially the quarter wave plate.

The alignment method used was that described by Aspnes and Studna (1971). They pointed out that the use of a transparent reflecting surface eliminated the effect of first order ellipticities in the polarizer and analyser. These effects can produce errors in the alignment. The procedure of a typical alignment was as follows.

The light source (He-Ne Laser) was adjusted with the analyzer arm in the straight-through position. With the quarter wave plate iris stopped down to its smallest position, and with no aperture in the analyzer arm, the direction of the laser beam was adjusted to give a maximum signal from the detector.

Next the angle of incidence zero error was checked. Apertures were inserted in both ends of the analyzer arm to define the axis of this arm. The analyzer arm was then moved in 0.01° steps about 90° and the angle of incidence was plotted vs. the detector signal. The resulting curve was symmetrical about its maximum and peaked at 89.97°, indicating an error of -0.03° in the scale (Fig. F.1).

The zero errors in the polarizer and analyzer scales were checked with the quarter wave plate removed, and with the angle of incidence at 70°. An optically flat quartz slab was used as a reflector.
Fig. F.1 Zero correction for the angle of incidence scale.

Fig. F.2 Corrections to the analyser and polarizer scales. (o-o-o) balancing the analyser near 0° for set values of P near 90° (outer scales for abscissa and ordinate). (x-x-x) balancing the polarizer near 0° for set values of A near 90° (inner scales for abscissa and ordinate). The correct correction factors are: \( A = -0.69°, P = -1.85° \).
The slab was thick enough to exclude the reflection from its near surface from entering the analyser arm. With both P and A set to approximately 90°, the position of the slab was adjusted for a maximum signal from the detector. This procedure ensures that the front surface of the reflector lies on the axis of rotation of the P and A arms, and that the surface around, and longitudinal axes of the P and A arms all lie in one plane, thus establishing the plane of incidence.

The analyser was then nulled near A = 0° for set values of P about 90° to obtain a straight line plot of A vs. P. Then the polarizer was nulled for P near 0° for set values of A about 90°, to obtain another straight line. These lines were plotted with a $-\pi/2$ shift in the ordinate and $+\pi/2$ shift in the abscissa of the second curve, as shown in Fig.F.2. At the point of intersection the transmission axes of the analyser and polarizer are either parallel or perpendicular to the plane of incidence. The correction in the two scales was made by setting the polarizer and analyser to the intersection values and then rotating the shaft encoders to read 90.00° or 0.00°.

To determine the zero error in the quarter wave plate scale, the polarizer was set to 0.00° and the analyser set to 90.00°. The quarter wave plate rotated about 0.00° to locate the position of minimum signal at the detector. When the signal is a minimum, the first axis of the quarter wave plate is parallel to the polarizer transmission axis and so the error in the quarter wave plate scale can be found.(Fig. F.3).
Fig. F.3 Zero correction for the quarter wave plate scale.
For all of the measurements made with the ellipsometer, the quarter wave plate was positioned at -45°. An ideal quarter wave plate would have a relative phase retardation, $\Delta_c$, of 90.00° and a transmittance ratio, $T_c$, of 1.0°. Small deviations from these values can produce large errors in the ellipsometry readings and so it is advisable to have as perfect a quarter wave plate as possible.

To adjust $\Delta_c$ and $T_c$, the analyser arm was fixed in the straight-through position with $P$ at 0.00°, $A$ at 90.00°, and the quarter wave plate at 315.00°. The tuning micrometer screw on the Soleil-Babinet compensator was turned and the positions which gave extinction were noted. These positions correspond to retardations of 0, $2\pi$, $4\pi$,... Quarter wave retardation (i.e. $n\pi/2$) is found by adding one-quarter of the difference between extinction settings to any one of the settings. The final setting of the quarter wave plate was accomplished using the ellipsometer readings of a carefully aligned, Inconel-coated glass slide. For a perfectly aligned instrument, the ellipsometry readings in zones 1 and 3 will give the same values of $\psi$ and $\Delta$.* The micrometer on the compensator was adjusted to minimize the spread in the $\psi$'s and $\Delta$'s. It was found that the position

*The relations between zones 1 and 3 are:

<table>
<thead>
<tr>
<th>Zone 1</th>
<th>$\Delta = 90 + 2P_1$</th>
<th>$135 &gt; P_1 &gt; -45$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\psi = A_1$</td>
<td>$90 &gt; A_1 &gt; 0$</td>
</tr>
<tr>
<td></td>
<td>$Q = -45$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Zone 3</th>
<th>$\Delta = 2P_3 - 90$</th>
<th>$225 &gt; P_3 &gt; 45$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\psi = 180 - A_3$</td>
<td>$180 &gt; A_3 &gt; 90$</td>
</tr>
</tbody>
</table>

McCrackin(1963) gives a description of all the zones.
of minimum difference in $\psi$ was not the same as the position of minimum difference in $\Delta$. A series of readings were taken for different positions and using the program of McCrackin (1969), $\Delta_c$ and $T_c$ were calculated. The setting chosen gave $\Delta_c = 90.077^0$ and $T_c = 0.99959$. 
APPENDIX G *

ELLIPSOMETRIC INVESTIGATION OF THE ELECTRO-OPTIC AND ELECTROSTRICTIVE
EFFECTS IN ANODIC Ta\textsubscript{2}O\textsubscript{5} FILMS (Cornish & Young 1973)

G. 1 Introduction

Ellipsometry may be used to detect and measure separately the small changes in refractive index and thickness which occur in thin dielectric films when a field is applied. The occurrence of birefringence is also detectable. The ellipsometric technique consists in measuring the relative phase and amplitude changes in the $s$ and $p$ components of light on reflection from the film. The $s$ and $p$ components have the electric vector perpendicular and parallel to the plane of incidence respectively. If $R_p$ and $R_s$ are the complex reflectivities for the two components, one obtains $\psi$ and $\Delta$ where

$$R_p/R_s = (\tan \psi) \exp(i\Delta).$$

In the present work, a PDP8-E computer was used to perform the ellipsometer balancing procedure as well as to record currents, voltages and elapsed time.

In previous work on these films, Holden and Ullman(1967,1969) discovered the modulation of intensity reflection on applying a.c. fields to the films using a monochromator with a lock-in amplifier detection technique. They explained their results in terms of thickness modulation. Frova and Migliorato(1968,1969) attributed the effect to refractive index changes and compared their coefficients to those of oxygen-octahedra ferroelectric crystals.

Ord, Hopper and Wang(1972) were the first to apply ellipsometry to this problem. They were able to show that the thickness increased and the index decreased on increasing an applied field. It had previously been mistakenly assumed that the films would become

* This appendix represents a continuation and refinement of work submitted for the M.A.Sc. degree and led to the publication listed. The method developed has since been applied to Nb\textsubscript{2}O\textsubscript{5} by Yee and Young(1975).
thinner. These authors reported their data on the basis of a linear dependence on field, since only a narrow range of field was used, such as to give film growth (ionic conduction) at a rate which could be tracked by their automated ellipsometer. They treated the effect on the basis of an isotropic index change but in unpublished work, Hopper and De Smet have considered the question of anisotropy with a field applied (M.A. Hopper, personal communication).

The effects in question are of possible interest in applications such as integrated optical circuits, where they could be used in fabricating modulators (Frova and Miglioato(1968,1969). They are also of interest since they give information on the structural changes in these films, for example, on the changes which lead to strong history effects in the ionic conduction process which occurs in film growth. Also, a quadratic electrostriction effect was suggested (Young 1963 ) as one source of the quadratic field terms in the activation energies for ionic motion (Young 1960 ).

It may reasonably be assumed that the films, which have normally been considered to be amorphous, consist of a disordered form of one of the varieties of crystalline Ta₂O₅ which Stephenson and Roth have recently investigated in a series of papers (e.g. 1971 ). These structures appear to be complicated sequences of herringboned chains of fused pentagonal bipyramids or distorted octahedral bipyramids. It has been supposed that the history effects in the ionic conduction process indicate both point defect concentration changes and general distortion of the lattice. However, there is controversy over the explanation of those effects (Dell'Oca, Pulfrey and Young 1972); Dignam 1972 ).
G.2 Experimental Procedures

The automated ellipsometer was similar in general design to that used by Ord et al. (1972). A Rudolf (type 43603-200E) ellipsometer was modified by the addition of stepping motor drives (I.M.C. Magnetic Corp. type 008-008) on the analyser and polarizer. Anti-backlash gears (W.M. Berg, Inc.) were used, with one motor step corresponding to a 0.01° rotation of the optical elements. Software acceleration of the motors was employed. One difference from the design of Ord et al. was that absolute, brush-like shaft encoders (Theta Instrument Company) were used to read the analyser and polarizer angles. After each ellipsometer balance, the changes in the polarizer and analyser angles as indicated by the shaft encoders were compared with the estimate of those angles as calculated by the computer by algebraically summing output pulses sent to the motors. An error warning was printed indicating any discrepancies, such as would occur, for example, if a motor did not respond properly to every pulse. A 1 mW He-Ne laser source at 632.8nm was used. A Soleil-Babinet compensator (Gaertner Corp.) was used as the quarter wave plate. A photomultiplier (type RCA 931, later RCA 8645) was used as detector. It was preceded by a narrow band optical filter, thus allowing ordinary room illumination. The interface to a PDP8-E computer was constructed using standard Digital Corp. components. It provided digital and analogue inputs, a clock and also relay controls to initiate current flow. Balance time was not minimized but was typically 2.2s. The ellipsometer was aligned using the method of McCrackin, Passaglia, Stromberg & Steinberg (1963).

In situ measurements were made with a triangular prism cell
made by joining optical glass flats with epoxy resin. The angle of incidence (63.46°) was fixed by the need for normal incidence on the cell faces. The solution was 0.2M sulphuric acid and was controlled at 298K. The cathode was platinized platinum. Tantalum specimens (Materials Research Corp.) were single crystal slices electropolished in 10% by volume 48% by mass HF in 98% by mass H₂SO₄.

Measurements were made at intervals in two zones for calibration purposes. In tracking, one zone only was used. Cell window errors were corrected for.

G.3 Results

Since the films are solid, and, also, for that matter, since they are attached to a solid surface, it is to be expected that they would become anisotropic (birefrigent) on applying a field. One would expect that the films would become uniaxial with the optic axis perpendicular to the film surface. Since the films are grown by the application of high fields - that is in the anisotropic state and, furthermore, since the growth process is itself directional, in that the ions travel normal to the film, one might even expect that, on removal of the field, some anisotropy of the same type might remain frozen-in. Despite the failure to detect structure in these films in diffraction experiments therefore, they need not be completely free of microstructure or be completely isotropic.

The ellipsometry angles Ψ, Δ for homogeneous isotropic, non-absorbing films follow a closed loop (at least, modulo 2π in Δ). With anisotropy of the expected kind, the refractive index would be represented by an optical indicatrix which was an ellipsoid of rotation
about the film normal. If so, the s-light responds to $n_o$ the radius of the circular principal section. The p light responds to $n_p$, where if $\phi_1$ is the angle of incidence and $n_\perp$ the index of the ambient, the angle of refraction $\phi_2$ is given by $n_\perp \sin \phi_1 = n_p \sin \phi_2$ with the indicatrix determining a relation between $\psi$ and $n_p$;

$$n_p \sin^2 \phi_2/n_o^2 + n_p \cos^2 \phi_2/n_o^2 = 1.$$ 

The reflectivities $R_s$ and $R_p$ are then calculable in the usual way except that different indices $n_o$ and $n_p$ are used. The result is that, for an anisotropic film, the data spiral either upwards or downwards (depending on the sign of the birefringence) in the $\psi, \Delta$ domain instead of following a closed loop. The problem of ellipsometry with anisotropic films was considered recently by Engelsen (1971).

Fig. G.1 shows the lower part of $\psi, \Delta$ domain for films which were grown successively to greater thicknesses. The ellipsometer was balanced during the growth process (with the field on) and again during periodic interruptions (with the field removed). With the field removed, the data lie on a single curve for successive cycles of growth (see legend for optical constants). The data for zero field fit a curve for an isotropic film with constants as shown in the figure caption. With the field on, the curve traces a higher path on each cycle. The data for constant applied field fit computed curves for an anisotropic film. Thus, the data are consistent with the films being isotropic, homogeneous and non-absorbing within some limits with the field off, but becoming markedly anisotropic (though still homogeneous through their thickness) when the field was on.
Fig. G.1 Lower part of $\psi, \Delta$ domain for increasing thickness of films on tantalum up to three cycles. Lower curve (dashed) is computed for an isotropic oxide and the experimental points are for zero field: $\blacklozenge$, first cycle; $\bullet$, second cycle; $\Delta$, third cycle. Upper three curves (solid lines) represent an anisotropic film with experimental points for field applied: $\square$, first cycle; $\bigcirc$, second cycle; $\triangle$, third cycle. Optical constants: thantalum $n - ik = 2.46 - 12.573$; isotropic oxide, $n = 2.195$; anisotropic oxide, $n_o - n_n = -0.090$; $n_e - n_n = \beta (n_o - n_n)$, with $\beta = 1.6$.

Fig. G.2 Upper part of $\psi, \Delta$ domain. Key as for Fig. G.1.
Films grown in the present electrolyte do not show detectably the effects seen for films grown in phosphoric acid which consist of two layers (Dell'Oca & Young 1970). The outer layer has a lower index due to incorporation of phosphate; this layer is believed to grow due to metal ion motion as opposed to oxygen ion motion. This effect, however, gives a quite different kind of spiralling of the $\psi, \Delta$ plot.

Fig. G.2 shows similar results for the top portion of the curves, confirming the behaviour demonstrated by Fig. G.1.

To proceed further it was assumed that if $n_e$ and $n_o$ are the changes produced by a field $E$ and if $n_n$ is the index of the oxide with no field applied, then

$$\beta = \frac{\Delta n_e}{\Delta n_o} = \frac{(n_n - n_e)}{(n_n - n_o)}$$

is a constant independent of $E$. The method of separately determining changes of thickness and of index is illustrated by Fig. G.3. This shows, on the $\psi, \Delta$ plane, contours of equal thickness and equal $n_o$, calculated for our angle of incidence and assuming $\beta = 1.6$ as determined by obtaining the best fit of computed curves and experimental data in Fig. G.1 and G.2. Also shown are experimental data for a given film to which a sequence of fields was applied up to $5.07 \times 10^2$ MV m$^{-1}$. The decrease in index and the increase in thickness can be read off in Fig. G.3 for each value of the field. In the previous work by Ord et al. (1972) the films were assumed isotropic, i.e. in calculating similar charts the value of $\beta$ was taken as 1. The data for a single film do not exclude such treatment. The anisotropy is detected and measured only by growing the film so as to cover more than one cycle.
Fig. G.3 Contours (---) of constant index $n_o$ (with $\beta = 1.6$) and contours (-----) of constant film thickness on tantalum with substrate constants as in Fig. G.1. The experimental points are for a film on tantalum with a range of field applied as shown and maintained until steady-state changes were established. The fields were such that negligible ion current was produced and hence negligible growth.
APPENDIX H

PROGRAMS USED TO CONTROL THE ELLIPSMETER SYSTEM

H.1 Introduction

There were seven main programs used to control the automated ellipsometer and to collect data. The programs were written in a combination of Fortran and assembly language to be compatible with the OS8 operating system used by the PDP-8E computer. Using the programs listed in Table H.1, the polarizer and analyser readings could be accumulated while the crystal was scanned through the probing beam. The data was stored on the Dectape Unit for output via the teletype or the incremental plotter. Table H.2 lists the subroutines used by the main programs.

The most frequently used programs were callable from a keyboard monitor program (SKEY.SV). The functions that could be performed from the keyboard monitor were (i) step the xy motors which moved the crystal through the probing beam; (ii) position the pen of the xy plotter; (iii) balance the ellipsometer; and (iv) scan the crystal horizontally and record the polarizer angle, analyser angle, and position of the crystal. After finishing a command, the program returned to the keyboard monitor.

Other programs (DIFFA.SV and DIFFP.SV) were used to subtract either the analyser or polarizer readings of two scans along the crystal and to plot the difference on the xy plotter. The two programs ANAL.SV and POLA.SV plotted the analyzer and polarizer readings from a single scan. DATA.SV was used to type out the contents of a file where the data from one scan was stored.
<table>
<thead>
<tr>
<th>MAIN PROGRAMS</th>
<th>SUBROUTINES CALLED</th>
<th>FUNCTION</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. SKEY.SV</td>
<td>SRUN, SBEN, SSTEP, SPLTXY</td>
<td>Keyboard monitor program</td>
</tr>
<tr>
<td></td>
<td>SXYPEN, SMOT2, SBE1, SRDA</td>
<td></td>
</tr>
<tr>
<td></td>
<td>SRPHOT, SREV, STEPN</td>
<td></td>
</tr>
<tr>
<td>2. PLOT.SV</td>
<td>SAXES</td>
<td>Draws axes and calculates scale factors for plotting</td>
</tr>
<tr>
<td>3. POLA.SV</td>
<td>Saxes, SDEGR, SPLTXY</td>
<td>Plots polarizer readings from one file and types results</td>
</tr>
<tr>
<td>4. ANAL.SV</td>
<td>Saxes, SDEGR, SPLTXY</td>
<td>Plots analyser readings from one file and types results</td>
</tr>
<tr>
<td>5. DIFFA.SV</td>
<td>Saxes, SDEGR, SPLTXY</td>
<td>Subtracts analyser readings from two files, plots and types the difference</td>
</tr>
<tr>
<td>6. DIFFP.SV</td>
<td>Saxes, SDEGR, SPLTXY</td>
<td>Subtracts polarizer readings from two files, plots and types the difference</td>
</tr>
<tr>
<td>7. DATA.SV</td>
<td>SDEGR</td>
<td>Types contents of a file on the teletype</td>
</tr>
</tbody>
</table>

Table H.1 Main programs used to collect data and control the automated ellipsometer.
<table>
<thead>
<tr>
<th>FILE</th>
<th>CALLING NAME</th>
<th>FUNCTION</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>SBE1</td>
<td>BE1</td>
</tr>
<tr>
<td>2.</td>
<td>STEPN</td>
<td>STEPN</td>
</tr>
<tr>
<td>3.</td>
<td>SREV</td>
<td>REV</td>
</tr>
<tr>
<td>4.</td>
<td>SSTEP</td>
<td>STEP</td>
</tr>
<tr>
<td>5.</td>
<td>SRUN</td>
<td>RUN</td>
</tr>
<tr>
<td>6.</td>
<td>SMOT2</td>
<td>MOT2</td>
</tr>
<tr>
<td>7.</td>
<td>SAXES</td>
<td>AXES(IYR, IXR, IYMUL, IXMUL)</td>
</tr>
<tr>
<td>8.</td>
<td>SBEN</td>
<td>BE(P, A)</td>
</tr>
<tr>
<td>9.</td>
<td>SDEGR</td>
<td>DEGR(IL, IH, ANGLES)</td>
</tr>
<tr>
<td>10.</td>
<td>SPLTXY</td>
<td>PLTXY(I, X, IY)</td>
</tr>
<tr>
<td>11.</td>
<td>SXYPEN</td>
<td>XYPEN</td>
</tr>
<tr>
<td>12.</td>
<td>SRDA</td>
<td>RDA(IL, IH, ANGLES)</td>
</tr>
<tr>
<td>FILE</td>
<td>CALLING NAME</td>
<td>FUNCTION</td>
</tr>
<tr>
<td>--------</td>
<td>---------------</td>
<td>---------------------------------------------------</td>
</tr>
<tr>
<td>13. SRPHOT</td>
<td>RPHOTO(ERR)</td>
<td>Reads the error signal from the detector of the ellipsometer</td>
</tr>
</tbody>
</table>

Table H.2 Subroutines used by the main programs listed in Table H.1
H.2 Main Programs

1. SKEY.SV

```
C KEYBOARD COMMAND PROGRAM
COMMON IDIP,III,IIAH,IIPL,IIPH
ST,CLA
READ(1,2)INST
2 FORMAT('18.2E')
IF(INST=133)1,3,4
3 CALL SEL
4 CONTINUE
   IF(INST-292)10,5,10
5 CALL MOT2
10 CONTINUE
   IF(INST=117)6,7,6
7 CALL RUN
6 CONTINUE
   IF(INST=133)9,8,9
8 CALL YYPEN
9 CONTINUE
5 JMP ST
END
```

2. POLF.SV

```
C DRAW AXES AND CALCULATE SCALE FACTORS
WRITE(1,1)
1 FORMAT('ZERO PLOTTER')
READ(1,2)IXR
2 FORMAT('Y RANGE IN DEGREES = 'I3)
READ(1,3)IXR
3 FORMAT('X RANGE = 'I3)
CALL AXES(IXR,IXP,IXMR,IXMXR)
CALL EXIT
END
```
3. POLA.SV

C PLOT POLARIZER READINGS FOUND IN FILE ENTERED
C SR OPTION: IF SR11=11 TYPE OUT RESULTS ALSO.
D I M E N S I O N I A 1 ( 2 0 0 ) , I A 2 ( 2 0 0 )
D I M E N S I O N D P ( 2 0 0 ) , I X ( 2 0 0 )
D P H = - 3 7 0 .
D P L = 3 7 0 .
R E A D ( 1 , 1 ) : F I L E 1
1 F O R M A T ( ' E N T E R 1 S T D A T A F I L E N A M E : ' , A 6 )
R E A D ( 1 , 3 ) : N
3 F O R M A T ( ' N O . O F D A T A L I V E S = ' , I 3 )
C X Y P L O T T E R I N I T I A L I Z A T I O N
W R I T E ( 1 , I A )
1 4 F O R M A T ( ' P L O T T E R R E A D Y ? P R E S S C O N T . , S R 1 1 = 0 1 F O R A X E S ' )
S H L T
I X A = 0
I Y A = 0
C A L L I O P E N ( ' D T A 1 ' , F I L E 1 )
R E A D ( 4 , 4 ) : I A 1 ( J ) , I A 2 ( J ) , I A 3 , I A 4 , I X ( J ) , I A 5 , J = 1 , N
4 F O R M A T ( C A R )
D O 6 J = 1 , N
I A L = I A 1 ( 1 )
I A H = I A 2 ( 1 )
C A L L D E F ( I A L , I A H , P A )
D P ( 1 ) = P A
I F ( D P H - D P ( 1 ) ) > 7 , 8 , 8
7 D P H = D P ( 1 )
8 C O N T I N U E
I F ( D P L - D P ( 1 ) ) > 5 , 5 , 5
5 D P L = D P ( 1 )
9 C O N T I N U E
W R I T E ( 1 , 1 5 ) : D P H , D P L
1 5 F O R M A T ( ' P L O T L I M I T S : Y H I G H = ' , F 6 . 2 )
R E A D ( 1 , 1 6 ) : F Y H
1 6 F O R M A T ( ' S E T Y H I G H = ' , F 6 . 2 , ' Y L O W = ' , F 6 . 2 )
S Y H = I F I X ( F Y H - D P H )
S Y L = I F I X ( D P L - F Y H )
S I X = I X ( N )
C P L O T A X E S A N D S E T S C A L E
A 3 = F L O A T ( I Y Y ) / 1 0 .
W R I T E ( 1 , 9 ) : A 3
C A L L A X E S ( I Y Y , I Y M , I Y M U )
I Y M U = F L O A T ( I Y M )
C S C A L E D A T A A N D P L O T P O I N T S
D O 1 0 I = 1 , N
I X = I X ( I ) + I X M U
P P = ( D P ( I ) - D P L ) * 1 0 . * I Y M U
I P P = I F I X ( P P )
I D Y = I D X - I X Y
I D X = I P P - I X A
I X A = I X Y
I Y A = I P P
1 0 C A L L P L T X Y ( I D X , I D Y )
W R I T E ( 1 , 1 1 )
1 1 F O R M A T ( ' T Y P E R E S U L T S ? Y E S : S R 1 1 = 0 1 : P R E S S C O N T ' )
S H L T
S C L A
S 7 1 0 4
S A N D ( X 1
S S 7 A C L A
S J M S T Y P E
C A L L E X I T
S T Y P E , 8
W R I T E ( 1 , 1 9 ) ( I X ( 1 ) , D P ( I ) , I = 1 , N )
1 9 F O R M A T ( I 3 , F 8 . 2 )
S J M P 1 T Y P E
E N D
PLOT ANALYSER READINGS FOUND IN FILE ENTERED

IF SR11=1; TYPE OUT RESULTS ALSO.

DIMENSION IA1C200),IASC00)
DIMENSION DPC00),IXC800)

DPH=-370.
DPL=370.

READ(1,1)FILE1

FORMAT('ENTER 1ST DATA FILE NAME: 'A6)

READ(1,3)N

FORMAT('NO. OF DATA LINES='I3)

XY PLOTTER INITIALIZATION

WRITE(1,14)

FORMAT('PLOTTER READY? PRESS CONT.. SR11=01 FOR AXES')

HLT.

IXA=0
IXY=0

CALL IOPEN('DATA1',FILE1).

READ(4,4)IA3,IA4,IA1(J),IAS(J),IX(J),IA5,J=1,N)

FORMAT(AA2)

DO 6 I=1,N

IAl=IA1(I)

IAH=IA2(I)

CALL DOPEN(IA1,IAH,PA)

DP(I)=3A0.-PA

IF(DPH-DC(I))7,8,8

DPH=DP(I)

CONTINUE

IF(DPL-DC(I))6,5J5

DPL=DC(I)

CONTINUE

WRITE(1,15)DPH,DPL

FORMAT('PLOT LIMITS: YHIGH='F6.2' YLOW='F6.2)

READ(1,1H)DPH,DPL

FORMAT('SET YHIGH='F6.2' YLOW='F6.2)

IYM=IFIX(DPH-DPL)

IXM=IX(N)

C PLOT AXES AND SET SCALE

A3=FLOAT(IYM)/18.

WRITE(1,9A3)

FORMAT('EACH Y DIVISION =F4.2' DEGREES')

CALL AXES(IYM,IXM,IXMUL,IXMUL)

YMUL=FLOAT(IYMUL)

C SCALE DATA AND PLOT POINTS

DO 10 I=1,N

IX=IX(I)*IXMUL

PP=CP(I)-DPL)*IAl*YMUL

IPP=IFIX(CP)

IDX=IX-IAX

IDY=IPP-IAY

IXA=IX

IYA=IPP

CALL PLTXY(IDX,IDY)

WRITE(1,11)

FORMAT('TYPE RESULTS? YES: SR11=01; PRESS CONT')

HLT.

S CLA

S 7496

S AND (K1

S S7A CLA

S JNS TYPE

CALL EXIT

S TYPE,0

WRITE(1,12)(IX(I),DPC(I),I=1,N)

12 FORMAT(I3,F8.2)

S JMP 1 TYPE

END
5. DIFFA.SV

C SURFACE AIRL. READINGS FOUND IN THE 3 FILES ENTERED.
C PLOT THE DIFFERENCE VS. DISTANCE ALONG CRYSTAL
C SP OPTION: IF SF11=1, TYPE OUT RESULTS ALSO.
D DIMENSION IA1(5AM), IA2(5AM), IA3(5AM), IA4(5AM)
D DIMENSION E(5AM), F(5AM)
N=1=770.
DPL=170.
FILE1
1 FORMAT('PLOT IST DATA FILE NAME: 'AA)
FILE2
2 FORMAT('PLOT 2ND DATA FILE NAME: 'AA)
FILE3
3 FORMAT('NO. OF DATA LINES=')
N
NY PLOTTER INITIALIZATION
WRITE(1,16)
16 FORMAT('PLOTTER READY? PRESS CONT. SF11=01 FOR AXES')
S IV=A=0
IVX=0
CALL IOPEN('DATA1','FILE1')
FILE1
A FORMAT('DATA2')
CALL IOPEN('DATA2',FILE2)
FILE2
B READ(A,8)(IA1,IA2,IA3,IA4),IV(J),IA5,IA6,N)
C READ(A,8)(IA1,IA2,IA3,IA4),IV(J),IA5,IA6,N)
D DIMENSION MAX AND MIN VALUES OF PP
DO 1=1,N
IMAX=IA1(J)
IMIN=IA1(J)
CALL IBGR(IAM,1AM,1AM)
1BAM=1AM
IC=IBAM(J)
CALL IBGR(IAM,1AM,1AM)
IC=IBAM(J)
DO 2=1,PP
WRITE(1,19)IC,J,PP(I)
2 CONTINUE
2 FORMAT('SET DEL A=FR.*2')
DO 3=1,N
PP=PP(I)
3 CONTINUE
3 FORMAT('PP=PP(11),17,8,8')
4 PP=PP(I)
4 CONTINUE
WRITE(1,18)PP,PP
18 FORMAT('PP=PP(11),17,8,8')
C PLOT AXES AND SET SCALE
A=FLOAT(WX)/10.
WRITE(I,19)A
19 FORMAT('A=FLOAT(WX)/10.')
B FORMAT('EACH Y DIVISION =FR.*2 DEGREES')
CALL AVES(WX,IX1,IX2,IX3,IX4)
C SCALE DATA AND PLOT POINTS
DO 5=1,N
IX=IX(J)*Y
5 CONTINUE
PDP=(PP(11)-PP)*1A.*Y
IPP=IFIX(PDP)
IXV=IXV-IX
IXV=IPPP-IXV
IXV=IXV
6. DIFFP.SV

C SUBTRACT POL. READINGS FOUND IN THE P FILES ENTERED
C PLOT THE DIFFERENCE VS. DISTANCE ALONG CRYSTAL
C SP OPTION: IF SP11=1: TYPE OUT RESULTS ALSO.
C DIMENSION IA1(255),IAA(255),IAB(255),IB1(255),IB2(255)
C DIMENSION DPH(255),X(255)
C DPH=375.
C READ(1,1)FILE1
C FORMAT('ENTER 1ST DATA FILE NAME: 'A6),READ(1,2)FILE2
C FORMAT('ENTER 2ND DATA FILE NAME: 'A6),READ(1,3)FILE3
C XY PLOTTER INITIALIZATION
C WRITE(1,14)
C FORMAT('PLOTTER READY? PRESS CONT. SR11=01 FOR AXES')
C HLT
C IXA=0
C IXY=0
C CALL IOPFN('DTA1',FILE1)
C READ(4,4)IA1(J),IAB(J),IA3,IA4,IX(J),IAS,J=1,N)
C FORMAT('A6'),CALL IOPFN('DTA1',FILE2)
C READ(4,4)IB1(J),IB2(J),IAB(J),IAS,IAS13,IB4,J=1,N)
C FIND MAX AND MIN VALUES OF DP
C DO 6 I=1,N
C IAL=IA1(I)
C IAH=IA2(I)
C CALL DEGR(IAL,IAH,PA)
C IBL=IB1(I)
C IBH=IB2(I)
C CALL DEGR(IBL,IBH,PB)
C DP(I)=PA-PB
C IF(DPH-DRP(I))(7,8,9
C DPH=DP(I)
C CONTINUE
C IF(DPL-DRP(I))(6,5,5
C DPL=DP(I)
C CONTINUE
C WRITE(1,15)DPH,DPL
C FORMAT('PLOT LIMITS: YHIGH='F6.2' YLOW='F6.2')
C READ(1,13)DPH,DPL
C FORMAT('SET YHIGH='F6.2' SET YLOW='F6.2')
C IXY=IFIN(DPH,DPL)
C IXY=IFY(N)
C PLOT AXES AND SET SCALE
A3=FLOAT(IXY)/19.
WRITE(1,9)A3
FORMAT('EACH Y DIVISION = 'F4.2 DEGREES')
CALL AXES(IYM,1YM,1YMUL,IXMUL)
YMU$=FLOAT(1YMUL)
SCALE DATA AND PLOT POINTS
DO 10 I=1,N
IXI=IX(I)*IXMUL
PP=(IPX(I)-DP)*10.*YMUL
IPP=IFIX(PP)
IX=IYI-IX
IP=IPP-IY
IXA=IX
IYA=IY
CALL PLTVX(IXM,1Y)
WRITE(1,11)
11 FORMAT('TYPE RESULTS? YES: SP11=311 PRESS CONT')
S
MLT
S
CLA
S
AND XI
S
STA CLA
S
JMS TYPE
CALL EXIT
S
TYPE=9
WRITE(1,19)(IX(I),IPX(I),I=1,N)
S
FORMAT('NO OF LINE IN FILE='I13)
READ(1,14)N1,N2
14 FORMAT('LIST LINES 'I3' TO 'I3')
CALL INPEM('DTAI',FNAME)
DO 3 I=1,N
READ(1,11)(IPuthor(I),IPX(I),I=1,N)
CONTINUE
3 CONTINUE
DO 2 I=1,N2
IA1=IPL(I)
IA2=IPH(I)
CALL DEGRI(IA1,IA2,ANG)
P(I)=ANG
IA1=IAL(I)
IA2=IAX(I)
CALL DEGRI(IA1,IA2,ANG)
A(I)=360.-ANG
CONTINUE
WRITE(1,18)
10 FORMAT(' P A Y X')
DO 11 I=N1,N2
WRITE(1,12)(A(I),IY(I),IX)
12 FORMAT(F6.2,1X,F6.2,1X,13,IX,13)
CONTINUE
CALL EXIT
END

7. DATA.SV

READ CONTENTS OF FILENAME AND TYPE ON TTY
ANGLES ARE CONVERTED TO DEGREES FOR OUTPUT
DIMENSION P(200),A(200)
DIMENSION IPLP(200),IPH(200),IAL(200),IAX(200),IY(200)
READ(1,13)FNAME
13 FORMAT('ENTER DATA FILE NAME='A6)
READ(1,11)N
11 FORMAT('NO. OF LINE IN FILE='I3)
READ(1,14)N1,N2
14 FORMAT('LIST LINES 'I3' TO 'I3')
CALL INPEM('DTAI',FNAME)
DO 3 I=1,N
READ(1,11)(IPuthor(I),IPX(I),I=1,N)
CONTINUE
3 CONTINUE
DO 2 I=1,N2
IA1=IPL(I)
IA2=IPH(I)
CALL DEGRI(IA1,IA2,ANG)
P(I)=ANG
IA1=IAL(I)
IA2=IAX(I)
CALL DEGRI(IA1,IA2,ANG)
A(I)=360.-ANG
CONTINUE
WRITE(1,18)
10 FORMAT('P A Y X')
DO 11 I=N1,N2
WRITE(1,12)(A(I),IY(I),IX)
12 FORMAT(F6.2,1X,F6.2,1X,13,IX,13)
CONTINUE
CALL EXIT
END
H.3 Subroutines Called by the Main Programs

1. SBEL

C BALANCE ELLIPSOmeter AND PRINT P&A
SUBROUTINE SBEL
CALL DEP(A)
WRITE(1,1)P,A
1 FORMAT('P='F6.2,'A='F6.2)
RETURN
END

2. STEPN

C STEP Y MOTOR N TIMES TO ACCOUNT FOR GEAR REDUCTION
SUBROUTINE STEPN
S CLA
DO 1 I=1,10
CALL STEP
S TAD WAIT
S DCA W
S MT, ISZ W
S JMP 'MT
1 CONTINUE
RETURN
S 'WAIT,5000
S W,
END

3. SREV

C REVERSE MOTOR DIRECTION
SUBROUTINE SREV
COMMON IDIR
S CLA
S TAD \IDIR
S AND CK5
S CMA
S AND CK5
S DCA CHDIR
S TAD \IDIR
S AND CK6772
S TAD CK1000
S TAD CHDIR
S 6332
S DCA \IDIR
RETURN
S CHDIR,0
END
STEP MOTOR ONCE

SUBROUTINE STEP
COMMON I, DIP, S, CLA, TAD, MDIR, AND CK1000
S7A CLA
JMP ACCST
TAD ACCSTP
S7A CLA
JMP ACCNTU
JMP STEP1
RETURN
ACCST, IAC
S7A
JMP ACCST
TAD CK1000
CMA
AND \VIDIR
DCA \VIDIR
TAD CK-10
DCA ACCSTP
TAD CK5700
DCA \TIME
ACCNTU, JMS STEP1
TAD \TIME
TAD CK1000
DCA \TIME
TAD \TIME
B, IAC
S7A
JMP B
1ST ACCSTP
RETURN
RETURN
ACCSTP, 0
\TIME, 0
STEP1, 0
6334
CLA
TAD \T
C, IAC
S7A
JMP C
JMP I STEP1
\T, 7000
END
SCAN THE CRYSTAL

COMMON IDP,IIAL,IIAH,IPL,IPH
DIMENSION IAL(100),IAH(100),IPL(100),IPH(100),IY(100)
READ(1,1)IX,IY
1 FORMAT('STARTING POSITION: X='I3,' Y='I3)
READ(1,2)INC1
2 FORMAT('1ST SEC: M='I3,' INC='I2)
READ(1,3)INC2
3 FORMAT('2ND SEC: M='I3,' INC='I2)
C XY PLOTTER INITIALIZATION
IXA=0
IYA=0
6563 READ(1,30)IXM,IXM
30 FORMAT('PLOTTER: IXM= 'I3,' IYM= 'I2)
READ(1,30)P1
33 FORMAT('SET Y-ZERO='F6.2)
READ(1,30)NAME
32 FORMAT('DATA STORAGE FILE (6 LETTER NAME)= 'A6)
WRITE(1,32)
31 FORMAT('SR AND PLOTTER SET? PRESS CONT')
S HLT
IYM=FLOAT(IYM)
CALL OPEN('DTA1',NAME)
K=100
J=1
CALL RE(P,A)
IAL(J)=I1AL
IAH(J)=I1AH
IPL(J)=I1PL
IPH(J)=I1PH
IY(J)=0
S JMS PLOT
N=N1
INC=INC1
S JMS SCAN
N=N2
INC=INC2
S JMS SCAN
N=N1
INC=INC1
S JMS SCAN
KK=J
S JMS TAPE
CALL OCLOSE
WRITE(1,10)IX,Y,P,A
10 FORMAT('FINISHED: X='I3,' Y='I3,' P='F6.2,' Y='F6.2/)
RETURN
S SCAN=0
DO 4 I=1,N
J=J+1
DO 7 I2=1,INC
S CLA
S TAD (K60)
6332
7 CALL STEP
IY=IY+INC
IY(J)=IY
CALL RE(P,A)
IAL(J)=I1AL
AHC(J) = IIAH
IPL(J) = IIPL
IPH(J) = IIPH
C      IS SP9=1, PLOT (I1Y, P)
S      7600
S      AND (K3)
S      STA CLA
S      JMS PLOT
C      IF SP11=1, WRITE P, A
C      IF SR10=1, HALT
S      CLA
S      7600
S      AND (K3)
S      DCA / ISA
C      IF(ISP) = 25, 25, 20
S      WRITE(1,21)P,A,IIY
S      FORMAT(2F7.2,14)
S      GO TO(25,23,23)ISP
S      CONTINUE
S      HLT
C      IF SR05=1, RETURN
S      7600
S      AND (K100)
S      STA CLA
S      RETURN
C      AFTER 100 READINGS, OUTPUT TO TAPE
S      IF(ISP=J),ISP,5,S
S      CONTINUE
S      JMS TAPE
S      CONTINUE
S      JMP I SCAN
S      TAPE, 0
S      DO 9 X = 1, KM
S      WRITE(A,R) IPL(K), IPH(K), IAL(K), IAH(K), IY(K), IIX
9      FORMAT(6A2)
S      JMP I SCAN
S      PLOT, 0
S      IYI = IY * IXMUL
S      PP = (P-P1) * 10. * YML
S      IPP = IFF(PP)
S      IDX = IYI - IMA
S      IDY = IPP - IYA
S      IYI = IYI
S      IYA = IPP
S      CALL PLTXY(IDX, IDY)
S      JMP I PLOT
END
MOVE X, Y MOTORS FROM THE KEY BOARD

SUBROUTINE MOT2

COMMON IDIR

IDIR=0

S
DCA "WAIT

READ(1,1)NX

READ(1,2)NY

2 FORMAT('X=','I4')

READ(1,3)NX

1 FORMAT('Y=','I4')

C MOVE Y MOTOR

IF (NY>3,4,5)

3 N=IABS(NY)

S CLA

S TAD CKAA

S JMS MOVEY

4 GO TO 6

5 N=NY

S CLA

S TAD CKAA

S JMS MOVEY

S MOVE X MOTOR

6 IF(NX>7,8,9)

7 N=IABS(NX)

S CLA

S TAD CKAA

S JMS MOVEY

8 RETURN

9 N=NX

S CLA

S TAD CK300

S JMS MOVEY

S RETURN

S MOVEX,0

S 6332

S CLA

DO 10 I=1,N

S CALL STEP

S 4T,15% "WAIT

S JMP QT

10 CONTINUE

S JMP I MOVE

S "WAIT,0

S MOVEY,0

S 6332

S CLA

DO 11 I=1,N

S CALL STEP

S JMP I MOVEY

END
7. SAXES

C DRAWS AXES AND CALCULATES SCALE FACTORS
SUBROUTINE AXES(IYR,IXR,IYML,IYML)

6500 CLA CLL
6506 IY=IY*10
IMAX=1000
IMAY=1750
IYML=IMAX/IXR
IYML=IMAY/IYP
WRITE(1,4)IXR,IYML,IYML

4 FORMAT('IYML=',I4,' IYML=',I4)
CLA
7604 ABD KK1
STA CLA
JMS AXIS
RETURN
AXIS,0
MODE=0
IP=IYR
IMUL=IYML
INC=IP*IYML/10
DO 10 I=1,10
DO 11 J=1,INC
TAD LDDNY
JMS XLATE
11 CONTINUE
DO 12 N=1,20
TAD LUDSY
JMS XLATE
12 CONTINUE
DO 13 X=1,20
TAD LDDUX
JMS XLATE
13 CONTINUE
DO 14 I=1,IP
TAD LUDSX
JMS XLATE
14 CONTINUE
IF(MODE)16,16,17
16 MODE=1
IP=IYR
IMUL=IXML
CLA
TAD LDEX
DCA LDDNY
TAD LUDSY
DCA LUDUX
TAD LDDUX
DCA LUDSX
DCA LDDUX
GOTO.15
15 CONTINUE
JMP I AXIS
LDEX,IP
LUDSY,11
LDDUX,6
LUDSY,5
LDDUX,22
LUDSY,21
LDDUX,42
LUDSY,41
XLATE,0
XA,6501
JMP YA
6506 CLA CLL
JMP I XLATE
INC=0
END
8. SBEN

C BALANCE ELLIPSOmeter ONCE.
SUBROUTINE BE (P,A)
COMMON IDIR,IIAL,IIAH,IIPL,IIPH
DIMENSION ES(20)
IBAL=0
31 IBAL=IBAL+1
CALL (K3)
JMS SETM
JMS BAL
CALL (K40)
GO TO(31,32)IBAL
32 CONTINUE
CALL RDA(IIAL,IIAH,A)
CALL RDA (IIPL,IIPH,P)
RETURN
C BALANCE, SPECIFIED UNIT
NSTEP=0
MINF=0
N=16
CONTINUE
SUM1=S
CONTINUE
SUM2=S
IF MINF=1: APPROACHING A MIN
3 IF(MINF=3,3,4
REVERSE MOTOR IF NOT APPROACHING A MIN
4 IF(SUM2-SUM1)5,5,6
6 CALL REV
MINF=1
GO TO 1
7 PASS€D THRU MIN? GO TO 7
5 SUM1=SUM2
GO TO 2
CONTINUE
SUM1=S
CALL REV
J=2*N
DO 8 I=1,J
CALL STEP
NSTEP=NSTEP+1
8 BUILD SUM2 UNTIL IT EQUALS SUM1
SUM2=0
DO 9 I=1,N
CALL PhDOTO(ERR)
CALL STEP
NSTEP=NSTEP+1
ES(I)=ERR
9 SUM2=SUM2+ERR
DROP LAST AND ADD NEW READING IN CIRCULAR BUFFER
12 I=1
10 CALL STEP
NSTEP=NSTEP+1
CALL RPHOTO(ERR)
SUM2=SUM2+ERR-ES(I)
ES(I)=ERR
IF(SUM2=SUM1)11,13,13
I=I+1
N1=N+1
IF(I-V1)10,12,12
GO TO 10
J=STEP/2
CALL RPHOTO(ERR)
DO 14  I = 1 , J
CALL STEP
JMP I BAL
S = SETM, 0
TAD (K100)
6332
DCA \\ILIR
JMP I SETM
SUM, 0
S = 0
DO 40 IS=1,N
CALL RPHOTO(ERR)
CALL STEP
S = S + ERR
JMP I SUM
END

9. SDEGR

CONVERT READINGS TO ANGLES
SUBROUTINE DEGR(IL, IH, ANGLE)
CLA
TAD I \IL
DCA ANGL
TAD I \IH
DCA ANGH
TAD ANGL
AND (K17
DCA \IA1
TAD ANGL
RTL
RTR
AND (K17
DCA \IA2
TAD ANGL
RTL
RTR
AND (K17
DCA \IA3
TAD ANGH
AND (K17
DCA \IA4
TAD ANGH
RTR
RTR
AND (K17
DCA \IA5
A = .01*FLOAT(IA1)+.1*FLOAT(IA2)+FLOAT(IA3)+
ANGLE=A+10.*FLOAT(IA4)+100.*FLOAT(IA5)
RETURN
ANGL, 0
ANGH, 0
END
10. SPLTXY

```
C PLOT ONE POINT
SUBROUTINE PLTXY(IX, IY)
  IDX = IX
  IDY = IY
  IF (IDX) 5, 6, 7
  CONTINUE
  PA, TAD LUDWX
  JMS XLATED
  ISZ \IDX
  JMP PA
  GO TO 6
  IDX = -IDX
  PB, TAD LUDFX
  JMS XLATED
  ISZ \IDX
  JMP PB
  CONTINUE
  IF (IDY) 8, 9, 10
  CONTINUE
  PC, TAD LUDSY
  JMS XLATED
  ISZ \IDY
  JMP PC
  GO TO 9
  IDY = -IDY
  PD, TAD LUDNY
  JMS XLATED
  ISZ \IDY
  JMP PD
  CONTINUE
  JMS WAIT
  6505
  JMS WAIT
  6503
  RETURN
  XLATE, 0
  XA, 6501
  JMP XA
  6506
  CLA CLL
  JMP I XLATED
  LUDWX, 05
  LUDFX, 11
  LUDSY, 41
  LUDNY, 21
  WAIT, 0
  XB, 6501
  JMP XB
  6502
  JMP I WAIT
END
```

11. SXYPEN

```
C MOUE THE XY PLTTER REV FROM TTY
SUBROUTINE SXYPEN
  READ(1, 1) IX, IY
  FOR WAIT('XINC = 'IA, 'YINC = 'IB)
  CALL PLTXY(IX, IY)
  RETURN
END
```
12. SRDA

READ THE SHAFT ENCODER SPECIFIED
SUBROUTINE RDA (IL, IH, ANGLE)

CLA CLL
6302 DCA ANGH
S DCA ANGL
S TAD ANGL
S .
S DCA \IA1
S TAD ANGL
S RTR
S RTR
S AND (K17
S DCA \IA2
S TAD ANGL
S RTL
S RTL
S RAL
S AND (K17
S DCA \IA3
S DCA \IA4
S TAD ANGL
S RTR
S RTR
S .
S DCA \IA5
A += 0.1*FLOAT(IA1)+.1*FLOAT(IA2)+FLOAT(IA3)
ANGLE= A+10.*FLOAT(IA4)+100.*FLOAT(IA5)
S TAD ANGL
S DCA I IL
S TAD ANGH
S DCA I IH
RETURN
S ANGL, 0
S ANGH, 0
END

13. SRPHOT

READ THE ERROR SIGNAL
SUBROUTINE SRPHOTO (ERR)

CLA
S TAD (K17
S 6323
S A, 6321
S JMP A
S 6324
S CMA
S DCA \IERP
ERR = FLOAT(IERR)
RETURN
END