MULTIPLE CHARGE CARRIER SPECIES AND THEIR EFFECTS IN
PHOTOREFRACTIVE TWO-BEAM COUPLING IN POTASSIUM NIOBATE

Thesis
Submitted to
The School of Engineering of the
UNIVERSITY OF DAYTON

In Partial Fulfillment of the Requirements for
The Degree of
Master of Science in Electro-Optics

By
Michael David Amonson
Dayton, Ohio
December, 2017
Name: Amonson, Michael David

APPROVED BY:

Dean R. Evans, Ph.D.  Partha P. Banerjee, Ph.D.
Advisory Committee Chairman  Advisory Committee Member
Principal Physicist  Professor, Department Chair
Materials & Manufacturing Directorate  Electro-Optics & Photonics Department
Air Force Research Laboratory

Joseph W. Haus, Ph.D.
Advisory Committee Member
Professor
Electro-Optics & Photonics Department

Robert J. Wilkens, Ph.D., P.E.
Associate Dean for Research and Innovation
Professor
School of Engineering

Eddy M. Rojas, Ph.D., M.A., P.E.
Dean, School of Engineering
ABSTRACT

MULTIPLE CHARGE CARRIER SPECIES AND THEIR EFFECTS IN
PHOTOREFRACTIVE TWO-BEAM COUPLING IN POTASSIUM NIOBATE

Name: Amonson, Michael David
University of Dayton

Advisor: Dr. Dean Evans

This thesis reports on an experiment to measure charge carrier contributions from different Fe species and their effects on beam coupling efficiency using self-pumped counter-propagating two-beam coupling in iron-doped potassium niobate KNbO₃:Fe. We used multiple continuous wave lasers operating across the visual spectrum to explore charge carrier creation from various transitions. Photorefractive grating formation data was acquired and analyzed using a new theoretical model which incorporates multiple charge carrier species. Initial analysis provides supporting evidence of a multiple charge carrier model and presents new insights about the effects of various charge carriers on the photorefractive periodic space-charge fields.
ACKNOWLEDGMENTS

Thank you to Dr. Dean Evans for leadership and advice in photorefractive physics and helping me to accomplish this thesis. I greatly appreciate Dr. Partha Banerjee’s help and assistance in modeling and theory. I value Dr. Joseph Haus’ guidance in writing and defending my thesis. Thanks to my wife Sarah and children Anastasia and Maxwell for their love & support throughout this endeavor. Additionally, thanks to Christopher Rosenbury, Manuel (Manny) Ferdinandus and Jennifer Reed for technical support.
TABLE OF CONTENTS

ABSTRACT ...................................................................................................................................................... iv
ACKNOWLEDGMENTS ........................................................................................................................................ v
LIST OF FIGURES ........................................................................................................................................... vii
LIST OF TABLES ............................................................................................................................................... ix
LIST OF ABBREVIATIONS ............................................................................................................................. x
LIST OF SYMBOLS ........................................................................................................................................ xi
CHAPTER 1 INTRODUCTION ........................................................................................................................ 1
  1.1 Overview ............................................................................................................................................. 1
  1.2 Background ......................................................................................................................................... 2
  1.3 Outline of Thesis ................................................................................................................................. 4

CHAPTER 2 EXPERIMENTAL INVESTIGATION OF COUNTER-PROPAGATING TWO-BEAM COUPLING ........................................................................... 5
  2.1 Laser System and Crystal Characteristics .......................................................................................... 5
  2.2 Experimental Setup/Design Overview ............................................................................................... 6

CHAPTER 3 THEORY & MODELING ............................................................................................................... 13
  3.1 Photorefractive Crystal Theory .......................................................................................................... 13

CHAPTER 4 BEAM COUPLING RESULTS & DISCUSSION ..................................................................... 21
  4.1 Measurement of $\Delta O D$ at different wavelengths ......................................................................... 21
  4.2 Interpretation of Results ..................................................................................................................... 29

CHAPTER 5 CONCLUSION & FUTURE WORK ............................................................................................. 37

REFERENCES .................................................................................................................................................. 39

APPENDIX A Mathematica Code ............................................................................................................. 42
APPENDIX B Established Method for Crystal Analysis ............................................................................ 52
APPENDIX C Matlab Code .......................................................................................................................... 54
APPENDIX D Thermodynamic Analysis of Retrapping Model ............................................................... 62
LIST OF FIGURES

Figure 1.1: Schematics of the energy levels of different defect centers (Fe$^{2+}$-V, Fe$^{3+}$-V and Fe$^{3+}$[Nb]) in the bandgap of KNbO$_3$ Left image: unreduced (as-grown) sample. Right image: reduced sample. ............................................................. 4

Figure 2.1: Diagram illustrating how the OPL changes when the beam enters the crystal ............................................................................................................................ 7

Figure 2.2: 532 nm laser beam at crystal stage with the aplanatic lens removed. Left image is before beam path optimization and image on the right is after. ................. 9

Figure 2.3: Top image is a schematic of the optical setup. Bottom images are photographs of the actual setup......................................................................................... 10

Figure 2.4: Telescoping shutter system ................................................................. 11

Figure 2.5: High frequency spatial interference in the beam. Image on the left is a superposition of interference from damaged PBSCs and back reflections off of low quality beam dumps. Image on the right is interference solely from the cheap beam dumps. ................................................................................................. 12

Figure 4.1: Absorption spectra for VLOC K83B2-3 KNbO$_3$:Fe crystal ....................... 24

Figure 4.2: Experimental data plots of 5 mW baseline. Exploded view on the right. ...... 26

Figure 4.3: Experimental data plots where excited charge carrier numbers are held constant. Exploded view on the right ....................................................................... 28

Figure 4.4: Single term curve fitting. A simple decaying exponential was used on the left and Cauchy-Lorentz on the right. Continued on the next page. ...................... 32

Figure 4.5: Single term curve fitting. A simple decaying exponential was used on the left and Cauchy-Lorentz on the right ............................................................ 33

Figure 4.6: Computer models simulating grating formation at 447 nm, 3 mW where A$_3$ and $\tau_3$ are allowed to vary. A$_3$ is on the left and $\tau_3$ is on the right. ................. 34

Figure 4.7: 447 nm data plots with two and three term curve fitting performed in the left and right images respectively ................................................................. 35
Figure 4.8: Data plots overlaid with curve fitting for maximum possible charge carriers at 523 nm and 532 nm. No evidence of hole competition is visible.................. 35

Figure 4.9: Left figure is the original data. Middle figure is the extrapolated curve. Right figure is original-extrapolation................................................................. 36
LIST OF TABLES

Table 2.1: Laser wavelengths and their associated f/# numbers ........................................... 6

Table 2.2: Crystal stage focal information for all lasers including beam waists and Rayleigh ranges .................................................................................................................. 9

Table 4.1: Absorption coefficients & power levels for method 2 ........................................ 23

Table 4.2: Absorption coefficients & power levels to excite the same number of charge carriers at each wavelength ......................................................................................... 23

Table 4.3: Important laser variables for accurately determining power levels in order to excite equal numbers of charge carriers ................................................................. 24

Table 4.4: Baseline curve fitting parameters. Minimal number of terms used for curve fitting ................................................................................................................................. 27

Table 4.5: Baseline curve fitting parameters. Maximum number of terms used for curve fitting ................................................................................................................................. 27

Table 4.6: Charge carrier numbers equal curve fitting parameters. Minimal number of terms used for curve fitting ................................................................. 28

Table 4.7: Charge carrier numbers equal curve fitting parameters. Maximum number of terms used for curve fitting ................................................................. 29
LIST OF ABBREVIATIONS

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>PR</td>
<td>photorefractive</td>
</tr>
<tr>
<td>EO</td>
<td>electro-optic</td>
</tr>
<tr>
<td>Fe</td>
<td>iron</td>
</tr>
<tr>
<td>eV</td>
<td>electron-volts</td>
</tr>
<tr>
<td>CW</td>
<td>continuous wave</td>
</tr>
<tr>
<td>OPL</td>
<td>optical path length</td>
</tr>
<tr>
<td>ND filter</td>
<td>neutral density filter</td>
</tr>
<tr>
<td>PBSC</td>
<td>polarizing-beam-splitter-cube</td>
</tr>
<tr>
<td>UV</td>
<td>ultraviolet radiation</td>
</tr>
<tr>
<td>Vis</td>
<td>visible light</td>
</tr>
<tr>
<td>NIR</td>
<td>near infrared radiation</td>
</tr>
<tr>
<td>f/#</td>
<td>focal length of the lens divided by beam waist diameter</td>
</tr>
<tr>
<td>CB</td>
<td>conduction band</td>
</tr>
<tr>
<td>C-L</td>
<td>Cauchy-Lorentz</td>
</tr>
<tr>
<td>HWHM</td>
<td>half width at half maximum</td>
</tr>
<tr>
<td>PDF</td>
<td>probability distribution function</td>
</tr>
</tbody>
</table>
LIST OF SYMBOLS

$\text{KNbO}_3$ potassium niobate

$\text{LiNbO}_3$ lithium niobate

$\text{Fe}^{2+}$ divalent iron atom

$\text{Fe}^{3+}$ trivalent iron atom

$\text{Fe}^{2+}[\text{K}]-\text{VK}$ $\text{Fe}^{2+}$ on $\text{K}^+$ site with an adjacent potassium vacancy at 0.85 eV

$\text{Fe}^{3+}[\text{K}]-\text{VK}$ $\text{Fe}^{3+}$ on $\text{K}^+$ site with an adjacent potassium vacancy at 2.35 eV

$\text{Fe}^{3+}[\text{Nb}]-\text{VO}$ $\text{Fe}^{3+}$ on $\text{Nb}^{5+}$ site with an adjacent oxygen vacancy at 2.54 eV

$n_a, n_b, n_c$ indices of refraction for each crystal axis

$\lambda$ wavelength

$L$ distance from the rear focal plane of the lens to the front face of the crystal

d distance from the front face of the crystal to the final focal position

$F$ lens focal length

$M^2$ Indicates how close a laser is to being a single mode TEM$_{00}$ beam, which in turn determines how small a beam waist can be focused. For a perfect Gaussian TEM$_{00}$ beam, the $M^2$ equals 1.

$W_0$ beam waist before entering the lens

$W_{0,L}$ beam waist after the lens

$z_R$ Rayleigh range
ΔOD  change in optical density
T  temperature, Kelvin
qe  electron charge, $1.60217657 \times 10^{-19}$ coulombs
τ  excited lifetime of charge carrier species before it is trapped in a dark fringe
$E_{sc}$  maximum attainable space-charge field in the experimental configuration
$\tau_{di}$  dielectric relaxation time
$N_D^+$  ionized donor density
ND  donor density
NA  acceptor density
NF  free electron density
s  photoexcitation coefficient
J  current density
I  intensity
$I_{dark}$  erasure intensity which corresponds to the grating erasure rate arising from the sum of thermal and optical erasure rates
$k_B$  Boltzmann's constant, $1.3806488 \times 10^{-23}$ m$^2$ kg s$^{-2}$ K$^{-1}$
µ  carrier mobility
$\gamma_r$  charge recombination coefficient
δ  photovoltaic transport length
$\varepsilon_s$  static permittivity
$\varepsilon_r^{eff}$  effective dielectric permittivity constant of KNbO$_3$
m  interference modulation depth
Is  signal intensity
**IP**  pump intensity

**ED**  Diffusion field. Electrons excited into the conduction band naturally move from the bright fringe areas where they were created to the dark fringe areas.

**Em**  Drift field. Related to the externally applied electric field. Movement of charge carriers, which is due to an applied electric field, often stated as the electromotive force over a given distance.

**E_0**  externally applied electric field

**EQ**  Saturation field. Maximum applied electric field beyond which electron diffusion velocity no longer increases.

**E_{PV}**  photovoltaic field

**φ**  phase

**ω**  angular frequency

**t**  Time

**k**  wave vector

**As**  amplitude of signal beam

**Ap**  amplitude of pump beam

**τ**  material response time

**Λ**  grating spacing

**K**  grating spacing “k” vector

**Γ**  gain coefficient/intrinsic coupling coefficient

**r_{eff}**  effective electro-optic coefficient

**r^{eff}_{ij}**  second rank electro-optic tensor

**r^{s}_{ijk}**  third rank clamped electro-optic tensor

**α**  linear absorption coefficient
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma$</td>
<td>absorption cross section per unit energy</td>
</tr>
<tr>
<td>$\hat{n}_p$</td>
<td>unit vector for pump beam polarization vector</td>
</tr>
<tr>
<td>$\hat{n}_s$</td>
<td>unit vector for signal beam polarization vector</td>
</tr>
<tr>
<td>$C_{ijkl}^E$</td>
<td>fourth rank elastic stiffness tensor</td>
</tr>
<tr>
<td>$e_{klj}$</td>
<td>third rank piezoelectric stress tensor</td>
</tr>
<tr>
<td>$\hat{n}_{i,j,k}$</td>
<td>unit K vector for the space-charge field</td>
</tr>
<tr>
<td>$\varepsilon_{ij}^s$</td>
<td>clamped dielectric tensor</td>
</tr>
<tr>
<td>$p_{ijkl}^E$</td>
<td>effective fourth rank elasto-optic tensor</td>
</tr>
</tbody>
</table>
CHAPTER 1
INTRODUCTION

1.1 Overview

Recent advances in photorefractive (PR) physics are opening the door to selectively tailored material characteristics. This work explores the fundamental physics occurring within iron-doped potassium niobate (KNbO₃:Fe). We utilized nonreciprocal light transmission through crystals of KNbO₃:Fe via PR beam coupling between counter-propagating beams formed from the incident beam and the Fresnel reflection from the rear face of the crystal. The direction of amplification within the crystal depends on the sign of the main charge carriers, the sign of the effective electro-optic coefficient, and the crystal orientation¹. Interference between the two counter-propagating beams excites mobile charge carriers in the bright fringes, which then diffuse to the dark regions. This gives rise to a periodic space-charge field. This field induces variations in the refractive indices through the linear electro-optic (EO) effect and a holographic reflection grating is formed²,³.

Following a well-established and reasonable model, such as the one proposed by Günter et al. for iron-doped lithium niobate (LiNbO₃:Fe),⁴ in the visible spectrum, Fe²⁺ ions provide a source of photoelectrons while Fe³⁺ ions serve as electron traps. This established model extends to cover KNbO₃:Fe as well. Nearly all researchers in the field
have exclusively used only one mobile charge carrier species\textsuperscript{4,5,6,7} to model and describe the creation of periodic space-charge electric fields (i.e. PR gratings). This thesis expands on the work of Basun and Evans\textsuperscript{8} wherein they have proposed and seen evidence of multiple charge carrier species playing a role in grating formation. In our research, we have seen evidence supporting the multiple charge carrier species model. In this thesis we discuss the behavior of electrons and holes as they form photorefractive gratings.

A hurdle to overcome when working with doped PR materials such as reduced KNbO\textsubscript{3}:Fe is the inhomogeneity or wide variations in beam coupling efficiency amongst crystals, even grown using the same apparatus\textsuperscript{1,9}. This inhomogeneity is common and contributes to uncertainty in separating the true underlying physics from things such as: unintentional impurities, and nonuniform dopant concentrations.

Domains arise from passing through multiple phase transitions during the cooling process after crystal growth is complete, but can easily be aligned by post-growth poling the KNbO\textsubscript{3} crystals at 180\textdegree C and applying 150 V/mm across the $c$-axis. This poling process only takes about one minute, then the crystal is slowly cooled down to room temperature before the voltage is turned off. This process develops uniform polarization throughout the crystal, resulting in a single crystal domain. This poling process does not appear to affect other material properties of the crystal.

1.2 Background

Perovskite-type oxygen octahedra ferroelectrics like KNbO\textsubscript{3} are increasingly important materials owing to their linear and nonlinear EO properties. The large nonlinear EO coefficients derive from the high packing density of oxygen octahedra\textsuperscript{10} and high spontaneous polarization at room temperature\textsuperscript{4,7,11,12}. Beyond typical uses of
KNbO$_3$ like frequency doubling, potential uses range from fast optical correlators used in optical computing$^4$ to image processing, image broadcasting, edge enhancement, and optical storage$^{13,14}$. In order to make these new applications feasible, a fundamental understanding of the physical phenomena involved in grating formation of doped KNbO$_3$ is necessary. This knowledge will enable the manufacturing and tailoring of more advanced KNbO$_3$ crystals.

In the past, many varieties of dopants have been used in KNbO$_3$ in attempts to increase desirable properties like PR responsivity and enhanced spectral sensitivity.$^{12}$ Of these dopants, iron has been one of the most promising impurities used in KNbO$_3$. When a large percentage of the Fe$^{3+}$ ions are reduced (by as much as a factor of 30 from the unreduced state$^8$) to Fe$^{2+}$ ions, then electrons become the dominant charge carrier species as opposed to holes in non-reduced crystals (see left and right images in Figure 1.1). This reduction also results in a tremendous increase in PR sensitivity and speed. However, it is not desirable to convert all Fe$^{3+}$ to Fe$^{2+}$, because traps are required for PR grating formation to occur. Building off the results discussed in reference 8, we assume that in reduced KNbO$_3$:Fe there are three dominant Fe charge carrier centers that can play a role in grating formation. The left side of Figure 1.1 shows the charge centers in unreduced KNbO$_3$. The right side illustrates how the charge centers change when the crystal is reduced. The reduced centers are:

(a) Fe$^{2+}$[K]-V$_K$ at $0.85\pm0.01$ eV (Fe$^{2+}$ on K$^+$ site with an adjacent potassium vacancy, hereafter referred to as Fe$^{2+}$-V) from which electrons are excited to the conduction band (photoionization).
(b) Fe\textsuperscript{3+}[K]-V\textsubscript{K} at 2.35 eV (hereafter referred to as Fe\textsuperscript{3+}-V), from which electrons can be excited from the valence band to a Fermi-level, leaving mobile holes in the valence band (charge transfer).

(c) Fe\textsuperscript{3+}[Nb] at 2.54 eV (Fe\textsuperscript{3+} on Nb\textsuperscript{5+} site, hereafter referred to as Fe\textsuperscript{3+}[Nb]) from which electrons can be excited to the conduction band (photoionization).

![Figure 1.1: Schematics of the energy levels of different defect centers (Fe\textsuperscript{2+}-V, Fe\textsuperscript{3+}-V and Fe\textsuperscript{3+}[Nb]) in the bandgap of KNbO\textsubscript{3}. Left image: unreduced (as-grown) sample. Right image: reduced sample.](image)

### 1.3 Outline of Thesis

In this work, we aim to thoroughly examine multiple charge carrier species and their roles in grating formation. Specifically, in Chapter 2, we discuss the equipment used and unique qualities of our experimental setup as well as obstacles and potential pitfalls that were overcome. In chapter 3 we review the underlying theories, models, and math involved in photorefractive beam coupling and suggest a possible new way to model multiple charge carrier species in computer simulations. Thereafter, in Chapter 4, we discuss data acquisition methods, their validity, and then examine the experimental results for new insights among which we see evidence of competing charge carriers during grating formation. Chapter 5 summarizes the results and suggests possible future work.
CHAPTER 2
EXPERIMENTAL INVESTIGATION OF COUNTER-PROPAGATING TWO-BEAM COUPLING

2.1 Laser System and Crystal Characteristics

Self-pumped counter-propagating two-beam coupling measurements were acquired using six continuous wave (CW) lasers; a 532 nm Coherent Verdi and five small diode-pumped solid-state (DPSS) lasers manufactured by CrystaLaser operating at 447, 523, 561, 633, 785 nm. A 5.25 mm X 5.25 mm X 5.25 mm x-y-z cut crystal of KNbO₃:Fe originating from boule K83B2-3 (VLOC) was carefully chosen for its relatively high and uniform PR properties. The crystal was grown with a Fe dopant concentration of 2000 ppm in the melt. The laser beams are focused onto the crystal using a +200 mm focal length aplanatic lens. The f/# for each beam is summarized in Table 2.1. f/# assumes a collimated beam and is defined as the focal length of the lens divided by the diameter of the beam when it enters the lens. In our experiment, the beams are polarized parallel to the crystal a-axis and propagation is along the c-axis. A Thorlabs DET36A photodiode is used to measure laser light transmission for a range of powers, which is interfaced to an oscilloscope for data acquisition.
Table 2.1: Laser wavelengths and their associated f/# numbers

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Beam diameter exiting the laser (mm)</th>
<th>Beam diameter at the crystal stage lens (mm)</th>
<th>f/#</th>
</tr>
</thead>
<tbody>
<tr>
<td>447</td>
<td>0.7</td>
<td>1.65</td>
<td>121</td>
</tr>
<tr>
<td>523</td>
<td>0.25</td>
<td>2.0</td>
<td>100</td>
</tr>
<tr>
<td>532</td>
<td>2.25</td>
<td>2.3</td>
<td>87</td>
</tr>
<tr>
<td>561</td>
<td>2.1</td>
<td>0.625</td>
<td>320</td>
</tr>
<tr>
<td>633</td>
<td>1.03</td>
<td>1.7</td>
<td>118</td>
</tr>
<tr>
<td>785</td>
<td>1.2</td>
<td>3.3</td>
<td>61</td>
</tr>
</tbody>
</table>

2.2 Experimental Setup/Design Overview

One can easily obtain two-beam coupling with nonuniform irregularly shaped beams without a precise knowledge of exactly where they are focusing inside the crystal. But future accurate transient solution modeling can be simplified and this experiment made repeatable by using a coherent Gaussian shaped beam, with accurate knowledge of the beam waist (i.e. focal spot) location at the crystal stage after passing through a lens. The ideal place to focus within a crystal depends on several factors, like the gain coefficient, the crystal thickness, and the f/#. The index of refraction for the a-axis of KNbO₃ is \( n_a = 2.2031 \) (valid for \( \lambda = 532 \) nm), which increases the converging optical path length (OPL). For example, if we find that the ideal place to focus inside a crystal is at 1 mm (\( OPL = 1 \ mm \)) inside the front face as seen in Figure 2.1, then we solve for \( \frac{OPL}{n_a} \approx 0.5 \ mm \). Assuming a perfectly collimated beam impinging upon a \( F = +200 \ mm \) lens, we find that the crystal face should be \( L = F - d = 199.5 \ mm \) from the lens.
The Rayleigh range is also important to know, because it indicates how precise we need to be in our focal point positioning. If the confocal parameter, which is two times the Rayleigh range, is on the same order as the crystal length, then precise positioning is unnecessary. The $\frac{1}{e^2}$ beam waist radius of the 532 nm Verdi laser is $W_0 = 1.15 \ mm$. $\frac{1}{e^2}$ refers to the distance from the beam axis where the irradiance has fallen to 13.5% of its central peak intensity. Nearly as important as the beam waist in determining Rayleigh range is the $M^2$ factor. $M^2$, or beam propagation ratio, is an indicator for how close a laser is to being a single mode TEM$_{00}$ beam. This determines how small of a beam waist is obtainable. A perfect Gaussian TEM$_{00}$ beam has an $M^2$ equal to 1.0.15 After passing through the lens and assuming a perfect $M^2$ of 1.0 the new waist diameter is calculated to be $2W_{0,L} = \frac{2M^2\lambda F}{\pi W_0} = 58.9 \ \mu m$. The Rayleigh range of this ideal beam is $z_R = \frac{\pi W_{0,L}^2}{M^2\lambda} = 5.1 \ mm$ and confocal parameter is 10.2 mm. As such, precision knowledge of the location of the focal point is not needed, because the beam is essentially collimated inside the entire crystal. From the equation above we see that as $M^2$ increases the Rayleigh range decreases. We obviously did not have ideal beams, so we spatially
profiled each laser to determine the $M^2$ and whether or not the Gaussian profiles were acceptable. As mentioned earlier, a good Gaussian profile is important for repeatability and simplified modeling. Table 2.2 shows the focused beam waist diameter and Rayleigh range for each wavelength using the actual $M^2$ value of 1.2.

Ideally all $f/\#s$ would be equal. But regardless, the fact that the $f/\#s$ are quite large means that the relative intensities for each spot size are in a comparable range and the large Rayleigh ranges mean that the beams are effectively collimated inside the crystal. Care was taken to not make the $f/\#$ too large nor too small. If the $f/\#$ is very large and the incoming power is very low, then the intensity of the Fresnel reflection off of the back surface, which leads to photoconductivity may not be high enough to overcome the dark conductivity (i.e. thermal activation of trapped charges)\textsuperscript{16} and no PR grating will form. Conversely, if the $f/\#$ is small, in the range of 10 or less, then the resulting Rayleigh range will be very small, bringing about a large beam divergence, leading to a low intensity Fresnel reflection and again no PR grating will form. Using a computer model to compare the effects of the different beam waists upon grating formation shows a difference in $\Delta OD$ of only 0.06 between the beam waists of 1.65 mm and 3.3 mm. If we compare the most extreme case of the smallest beam waist 0.63 mm to the largest 3.3 mm, the difference in $\Delta OD$ is still only 0.4. The 0.63 mm beam waist belongs to the 561 nm laser, which not only has power fluctuations, but also the unusual characteristic of a converging beam exiting the laser. In light of these issues, we weigh the 561 nm data very lightly and avoided using it to draw firm conclusions. Both the 0.06 and 0.4 variances in $\Delta OD$ can easily be swallowed up in the inhomogeneity of the KNbO$_3$:Fe crystal. Thus our comparisons and conclusions are built off of valid and comparable data.
Table 2.2: Crystal stage focal information for all lasers including beam waists and Rayleigh ranges

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Beam Diameter before Lens (mm)</th>
<th>f/#</th>
<th>Focused Beam Waist Diameter (µm)</th>
<th>Rayleigh Range (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>447</td>
<td>1.65</td>
<td>121</td>
<td>82.71</td>
<td>10.02</td>
</tr>
<tr>
<td>523</td>
<td>2.0</td>
<td>100</td>
<td>79.86</td>
<td>7.98</td>
</tr>
<tr>
<td>532</td>
<td>2.3</td>
<td>87</td>
<td>70.78</td>
<td>6.14</td>
</tr>
<tr>
<td>561</td>
<td>0.63</td>
<td>317</td>
<td>256.02</td>
<td>76.48</td>
</tr>
<tr>
<td>633</td>
<td>1.7</td>
<td>118</td>
<td>113.61</td>
<td>13.34</td>
</tr>
<tr>
<td>785</td>
<td>3.3</td>
<td>61</td>
<td>72.68</td>
<td>4.4</td>
</tr>
</tbody>
</table>

A Cinogy laser beam analyzer was used to characterize and improve the laser beam after each optical element and to remove stray reflections in the optical beam path. This allowed us to improve the initial M² of the beam just before the crystal stage aplanatic lens from ~1.8 to ~1.2.

![532 nm laser beam at crystal stage with the aplanatic lens removed. Left image is before beam path optimization and image on the right is after.](image1)

Additionally, this analyzer has been used to verify the exact focal position at the crystal stage. The optical path was optimized using the 532 nm 10W Coherent Verdi laser and accordingly the beam had to be greatly attenuated using optical wedges and ND filters to avoid damaging the CCD array inside the Cinogy analyzer. A final version of the optical setup is shown in Figure 2.3 below.
A telescoping shutter system (see Figure 2.4) was integrated into the optical beam path and an oscilloscope was used to optimize the time for full beam power on/off at the crystal stage to around 12 microseconds. This was important because KNbO₃:Fe begins to form an internal space-charge field at low power levels. If power ramps up slowly as the shutter opens, non-uniform grating formation will occur. The minimum 1/e time threshold for two-beam coupling is estimated to be around 50 microseconds for full-power on or off. Additionally, a beam collimation tester has been used to obtain a highly collimated beam after passing through the telescoping shutter system.
Shutters and lenses were repositioned and adjusted while back reflections were also eliminated. High frequency components causing interference patterns were observed as seen in Figure 2.5, which degraded the Gaussian laser beam quality. The source of this degradation was determined to be two damaged polarizing-beam-splitter-cubes (PBSC) used in conjunction with a half-wave plate to attenuate the beam. New high-power PBSCs were installed and the beam profile improved, but significant interference patterns were still observed. It was discovered that several of the beam dumps used at the PBSCs were inadequate to handle the amount of power dumped into them and reflected a significant portion of light, inducing the interference as seen in Figure 2.5 and causing focusing problems. Several highly efficient Black Hole beam dumps were installed in place of the original beam dumps, effectively eliminating the maladaptive back reflections at both PBSCs.
Figure 2.5: High frequency spatial interference in the beam. Image on the left is a superposition of interference from damaged PBSCs and back reflections off of low quality beam dumps. Image on the right is interference solely from the cheap beam dumps.

Understanding the intricacies, strengths, and weaknesses of the experimental setup and equipment is necessary to obtain useful ΔOD measurements. Of similar importance is understanding the physics model behind PR beam coupling. This understanding will facilitate an accurate interpretation of acquired data.
CHAPTER 3
THEORY & MODELING

3.1 Photorefractive Crystal Theory

In order to better understand the physics and mechanics of PR two-beam coupling within doped KNbO$_3$ a thorough examination of Kukhtarev’s material equations$^{17,18}$ as seen in (3.1-1) – (3.1-4) is required. This knowledge and understanding facilitate a more accurate interpretation of data and results as discussed in chapter 4. Our theoretical model is constructed off of established theory modified to incorporate multiple sources of charge carriers, including both holes and electrons. Additionally, this awareness provides insight and guidance into better computational simulations. Kukhtarev’s equations are expressed as,

Continuity Eqn.  \[ \frac{\partial N_F}{\partial t} = \frac{\partial N_D^+}{\partial t} + \frac{1}{q_e} \frac{\partial J}{\partial z} \]  (3.1-1)

Rate Eqn.  \[ \frac{\partial N_D^+}{\partial t} = (N_D - N_F^+)[s(I + I_{dark})] - \gamma R N_D^+ N_F \]  (3.1-2)

Current Eqn.  \[ J = q_e \mu N_F E_{sc} + \mu k_B T \frac{\partial N_F}{\partial z} + s I (N_D - N_D^+) q_e \delta \]  (3.1-3)

Gauss’s Law  \[ \varepsilon_s \frac{\partial E_{sc}}{\partial z} = q_e (N_D^+ - N_A - N_F) \]  (3.1-4)
Here, $N_F, N_D^+, N_D, N_A$, are the densities of the free electrons, ionized donors, donors ($\text{Fe}^{2+}+\text{Fe}^{3+}$), and acceptors ($\text{Fe}^{3+}$) respectively. $q_e$ is the charge of an electron, $J$ is the current density, $s$ is photo-excitation coefficient, $T$ is the absolute temperature, $k_B$ is Boltzmann’s constant, $\delta$ is the photovoltaic transport length, $\varepsilon_s$ is the static permittivity, $\gamma_r$ is the recombination coefficient, and $\mu$ is the mobility, $I_{dark}$ is the erasure intensity corresponding to the grating erasure rate arising from the sum of thermal and optical erasure rates.\(^{19}\) No analytical solution exists to Kukhtarev’s equations, but if we assume a small depth of fringe modulation we can express the growth of the space charge field amplitude $E_{sc}$ as,

\[
\frac{a}{\tau_{dl}} \frac{\partial E_{sc}}{\partial t} + bE_{sc} + cm = 0 \tag{3.1-5}
\]

where $E_{sc}$ is the maximum attainable space-charge field in the particular experimental configuration.\(^{12}\) $m$ is the interference modulation depth and $\tau_{dl}$ is the dielectric relaxation time.

\[
m = \frac{\sqrt{l_s l_P}}{l_s + l_P + l_{dark}} e^{-i\varphi}, \tag{3.1-6}
\]

\[
a = 1 + \frac{E_D}{E_M} - i \frac{E_0}{E_M}, \tag{3.1-7}
\]

\[
b = 1 + \frac{E_D}{E_Q} - i \left( \frac{E_0}{E_Q} + \left[ \frac{N_A}{N_D} \right] \left[ \frac{E_{pv}}{E_Q} \right] \right), \tag{3.1-8}
\]

\[
c = E_0 + E_{pv} + iE_D. \tag{3.1-9}
\]

Here $E_D = \frac{2\pi k_B T}{q_e \lambda}$ is the diffusion field, $\lambda = \frac{\lambda}{2\pi \sin(90)}$ is the grating spacing, $E_Q = \frac{q_e N_A \lambda}{2\pi \varepsilon_s}$ is the saturation field, $E_{pv}$ is the photovoltaic field, $E_0$ is any externally applied electric field, $E_M = \frac{\gamma_r N_A \lambda}{2\pi \mu}$ is the drift field. The space charge field modifies the crystal
refractive index through the electro-optic effect leading to a holographic grating which couples the pump and signal beams.\(^1\) The electric fields of the pump and signal beam can be expressed as,

\[
E_s(z, t) = \frac{1}{2} A_s(z, t)e^{i(-kz-\omega t)} + c. c. \tag{3.1-10}
\]

\[
E_p(z, t) = \frac{1}{2} A_p(z, t)e^{i(+kz-\omega t)} + c. c. \tag{3.1-11}
\]

\(k\) is the propagation constant in the medium and \(\omega\) is the angular frequency. The standing intensity fringes in the crystal caused by interference between these two beam is,

\[
I(z, t) = \frac{A_sA_p^*}{l_s+l_p} e^{2ikz} + c. c. \tag{3.1-12}
\]

Now to obtain the crystal’s material response we substitute eqn (3.1-8) into Kukhtarev’s equations and find an analytical solution to eqn (3.1-5). Solving for time, we find the following expression for the transient state electric space charge field:

\[
E_{sc}(t) = E_{sc}(0)e^{-bt\alpha^2} + \frac{cm(z)}{b} \left( e^{-bt\alpha^2} - 1 \right). \tag{3.1-13}
\]

In the steady state this can be simplified to

\[
E_{sc}(z) = \frac{-cm(z)}{b} \tag{3.1-14}
\]

The maximum \(E_{sc}\) occurs when the phase \(\varphi\), inside \(m\) eqn (3.1-6), is an integer multiple of \(2\pi\) making the exponential 1. The \(\sqrt{\frac{l_s l_p}{l_s+l_p+l_t}}\) part of \(m\) is then normalized. The space charge field can then summarily be written as,

\[
E_{sc}(z) = \frac{(E_0+iE_D+E_{pv})m(z)}{1+iE_D+iE_{pv}-\left( \frac{E_0}{E_0} + \frac{N_A}{N_A} + \frac{E_{pv}}{E_{pv}} \right)}. \tag{3.1-15}
\]
The related material response time is

\[ \tau = \tau_{di} Re \left( \frac{1 + \frac{E_D}{E_M} - i \frac{E_0}{E_M}}{1 + \frac{E_D}{E_Q} - i \left( \frac{E_0}{E_Q} + \left[ \frac{N_A}{N_D} \right] \frac{E_{pv}}{E_Q} \right)} \right) \]  (3.1-16)

The photovoltaic field \( E_{pv} \) within KNbO\(_3\), is considered to be negligible.\(^{20,21}\) \( E_0 \) is any externally applied electric field and in our case is zero. Knowing that \( E_0 = 0 \) and \( E_{pv} \approx 0 \) greatly simplifies \( E_{sc} \) which can be written as,

\[ E_{sc}(z) = \frac{iE_D m(z)}{1 + \frac{E_D}{E_Q}}. \]  (3.1-17)

The intrinsic gain coupling coefficient \( \Gamma \) can be expressed as,

\[ \Gamma = \frac{2\pi n^3 r_{eff} \cos(2\theta)}{\lambda \cos(\theta)} Im(E_{sc}). \]  (3.1-18)

Where \( n \) is the crystal's refractive index along the plane of beam polarization and \( r_{eff} \) is the effective electro-optic coefficient. The direction of power transfer depends on the sign of \( r_{eff} \) which in turn depends on the direction of propagation relative to the crystal c-axis. This is the source of unidirectional gain in counter-propagation two beam coupling.\(^{22}\) This research only examined incident beams normal to the surface of the crystal with large $f/\#s$, so $\theta = 0$, but for completeness we include the more general tensorial forms, which are necessary for other crystal angles and small $f/\#s$. \( r_{eff} \) is the scalar electro-optic coefficient and comes from a second rank electro-optic tensor \( r_{ij}^{eff} \)

\[ r_{eff} = \hat{n}_p \cdot r_{ij}^{eff} \cdot \hat{n}_s \]  (3.1-21)

Where \( \hat{n}_p \) and \( \hat{n}_s \) are the unit vectors for the pump and signal beam polarization vectors.

This leads to
\[ r_{ij}^{\text{eff}} = r_{ijk}^s \hat{n}_k + p_{ijkl}^E \hat{n}_l A_{km}^{-1} B_m \]  
(3.1-22)

Wherein

\[ A_{ik} = C_{ijkl}^E \hat{n}_l \hat{n}_l \quad B_i = e_{klj} \hat{n}_k \hat{n}_j \]  
(3.1-23)

\( r_{ijk}^s \) is the third rank clamped electro-optic tensor, \( p_{ijkl}^E \) is the effective fourth rank elasto-optic tensor, \( C_{ijkl}^E \) is the fourth rank elastic stiffness tensor, \( e_{klj} \) is the third rank piezoelectric stress tensor, and \( \hat{n}_{l,i,j,k} \) is the unit \( \mathbf{K} \) vector for the space-charge field.

The piezoelectric and electro-optic tensors influence the dielectric constant \( \varepsilon_s = \varepsilon_r^{\text{eff}} \varepsilon_0 \)

\[ \varepsilon_r^{\text{eff}} = \varepsilon_{ij}^s \hat{n}_i \hat{n}_j + \frac{e_{ijkl}^s \hat{n}_l \hat{n}_m \hat{n}_n A_{kl}^{-1}}{\varepsilon_0} \equiv \varepsilon_{\text{eff}} \]  
(3.1-24)

\( \varepsilon_{ij}^s \) is the clamped dielectric tensor. For light polarized in the crystal’s a-c plane, \( \varepsilon_{\text{eff}} \) can be estimated by the index ellipsoid,

\[ n(\theta) = \left( \frac{\cos^2 \theta}{n_a^2} + \frac{\sin^2 \theta}{n_c^2} \right)^{-1/2} = n_{\text{eff}}. \]  
(3.1-25)

Where,

\[ \varepsilon_{\text{eff}} = n_{\text{eff}}^2, \]  
(3.1-26)

which leads to,

\[ \varepsilon_{\text{eff}}^2 = \left( \frac{\cos^2 \theta}{n_a^2} + \frac{\sin^2 \theta}{n_c^2} \right)^{-2}. \]  
(3.1-27)

The full expansion of \( r_{\text{eff}} \) from eqn (3.1-21) with \( s \) polarization along the a-c plane is,
\[ r_{\text{eff}} = \frac{n_0^4 r_{13} \cos^3 \theta - 2n_0^2 n_2^2 r_{15} \sin^2 \theta \cos \theta + n_0^2 r_{23} \sin^2 \theta \cos \theta}{\varepsilon_{\text{eff}}^2}, \]  
(3.1-28)

and can now be expressed as,

\[ r_{\text{eff}} = \frac{n_0^4 r_{13} \cos^3 \theta - 2n_0^2 n_2^2 r_{15} \sin^2 \theta \cos \theta + n_0^2 r_{23} \sin^2 \theta \cos \theta}{\left(\frac{\cos^2 \theta + \sin^2 \theta - z}{n_0^2 - n_2^2}\right)^2}. \]  
(3.1-29)

As mentioned earlier, for this experiment \( \theta = 0 \), so \( r_{\text{eff}} \) and \( \Gamma \) simplify to

\[ r_{\text{eff}} = r_{13}, \]  
(3.1-30)

\[ \Gamma = \left[\frac{2m^3 r_{\text{eff}}}{\lambda}\right] \text{Im}(E_{sc}). \]  
(3.1-31)

From all of this one may now write the traditional expressions for the coupled equations for Gaussian beam counter-propagation two-beam coupling\textsuperscript{23}:

\[ \frac{\partial l_{\text{pump}}}{\partial z} = -\alpha l_{\text{pump}} - \Gamma \frac{l_{\text{pump}} l_{\text{signal}}}{l_{\text{pump}} + l_{\text{signal}} + l_{\text{Dark}}} - \frac{2(z-f)l_{\text{pump}}}{z_R^2 + (z-f)^2}, \]  
(3.1-32)

\[ \frac{\partial l_{\text{signal}}}{\partial z} = +\alpha l_{\text{signal}} - \Gamma \frac{l_{\text{pump}} l_{\text{signal}}}{l_{\text{pump}} + l_{\text{signal}} + l_{\text{Dark}}} - \frac{2(z-f)l_{\text{signal}}}{z_R^2 + (z-f)^2}. \]  
(3.1-33)

The Gaussian component of equations (3.1-32) and (3.1-33) is \( \frac{dl}{dz} = \frac{-2(z-f)l_{\text{pump}}}{z_R^2 + (z-f)^2} \) and \( \alpha \) is the internal absorption coefficient. The results in 4.2 need to be viewed through the lens of a new theoretical model as the established models do not seem to represent the complex physics at play. It is believed that the simple model can be made more accurate by including the contributions of different charge carriers through summing

\[ \Gamma \frac{l_{\text{pump}} l_{\text{signal}}}{l_{\text{pump}} + l_{\text{signal}} + l_{\text{Dark}}} \]  

terms for each charge carrier to obtain, \( \Gamma_{\text{eff}} = -\Gamma_{e1} + \Gamma_{\text{hole}} - \Gamma_{e2} \).

The new coupled equations are expressed as

\[ \frac{\partial l_{\text{pump}}}{\partial z} = -\alpha l_{\text{pump}} + \Gamma_{\text{eff}} \frac{l_{\text{pump}} l_{\text{signal}}}{l_{\text{pump}} + l_{\text{signal}} + l_{\text{Dark}}} - \frac{2(z-f)l_{\text{pump}}}{z_R^2 + (z-f)^2}, \]  
(3.1-34)
\[
\frac{\partial I_{\text{signal}}}{\partial z} = +\alpha I_{\text{signal}} + \Gamma_{\text{eff}} \frac{I_{\text{pump}} I_{\text{signal}}}{I_{\text{pump}} + I_{\text{signal}} + I_{\text{dark}}} - \frac{2(z-f)I_{\text{signal}}}{z_f^2 + (z-f)^2}. \quad (3.1-35)
\]

The key to understanding how the individual charge carriers influence grating formation lies within \(\Gamma_{\text{eff}}\). Each charge carrier has its own unique \(\Gamma\) which in turn has its own unique electric space charge field \(E_{\text{sc}}\). Within \(E_{\text{sc}}\), the saturation field \(E_Q\) will change through the dielectric constant \(\varepsilon_s\), donor density \(N_D\), and trap density \(N_A\).

Attempts to create an accurate transient state computer model using the modifications illustrated in equations (3.1-34) and (3.1-35), which would allow us to simulate the effects of the multiple charge species on grating formation, were not entirely successful and would be an appropriate endeavor for future work. Nevertheless, the physical and mathematical insight gained through attempting to make our theoretical model a computational model has broadened and improved our interpretation of the acquired data.

Equations (3.1-34) and (3.1-35) have been used in chapter 4 to calculate theoretical \(\Delta \text{ODs}\). As an example of how this was done, we will briefly walk through some of the common and unique values used to calculate the theoretical \(\Delta \text{ODs}\) within our computational program (see Appendix A). This program only accounts for one charge carrier at a time. Values shared among all wavelengths are: the lens focal length \(F\), the effective electro-optic coefficient \(r_{\text{eff}}\), the temperature \(T\), the position of the crystal \(a\), and the thickness of the crystal \(L\). Unique to each wavelength are the following values: beam waist radius \(W_0\), laser power \(P_{\text{in}}\), and the absorption coefficient \(\alpha\). Although \(\Gamma_{\text{eff}}, N_D, \text{and} N_A\) are different and unique for each wavelength, we do not yet know how to accurately predict the changes. It is possible to manually change \(\Gamma\) to bring the theoretical \(\Delta \text{ODs}\) within the experimental range. For example, to get the theoretical \(\Delta \text{OD}\) in the experimental range for 523 nm, set \(\Gamma = 30.8 \text{ cm}^{-1}\) to obtain \(\Delta \text{OD} = 1.55\). This
accounts for the competing holes (see Figure 1.1) in grating formation. Additionally, to get the theoretical ΔOD in the experimental range for 447 nm, set Γ=32.8 cm\(^{-1}\) to obtain ΔOD = 1.79. This accounts for the competing holes and the second electron species.

In calculating theoretical ΔODs in Table 4.3, we set \(\Gamma = 49.7 \, cm^{-1}\), \(N_D = 5 \times 10^{17} \, cm^{-1}\), and \(N_A = 1.21 \times 10^{23} \, cm^{-1}\) and do not change them. This would be appropriate and accurate if only one charge carrier were present. As an example of how this was done in Table 4.3, look at 532 nm. We set \(W_0 = 0.115 \, cm\), \(P_{in} = 5 \, mW\), \(\alpha = 1.91 \, cm^{-1}\) and run these unique values with the common values through the program to obtain ΔOD = 2.04. The large discrepancy seen between theoretical and experimental ΔODs clearly illustrates how the single charge carrier model breaks down in the real world. This is clear evidence of multiple charge carrier species participating in grating formation.
4.1 Measurement of ΔOD at different wavelengths

The optimized optical beam path and a solid understanding of the underlying PR physics enable the acquisition of accurate and insightful ΔOD data, allowing us to study the multiple charge carrier species’ properties. ΔOD was found by using the well-known formula $\Delta OD = \log\left(\frac{V_{\text{max}}}{V_{\text{min}}}\right)$ and has been calculated using the following method (see Appendix B for more details). The first step involved choosing a time length sufficient to capture full grating formation and some degree of the crystal grating’s steady state, but short enough to capture the fine details in the grating formation. $V_{\text{max}}$ was found at the highest point in this first oscilloscope data window. The next step was to capture the photorefractive grating’s steady state at approximately 1 minute after formation, which was long enough to ensure that only steady stage data was captured. The y-axis voltage data points at steady state were then averaged and used as $V_{\text{min}}$. Significant variances have been observed for each area studied on the KNbO3:Fe crystal. All experimental ΔOD values were calculated using Matlab (see Appendix C) and are listed in Table 4.4 – 4.7.

A minor complexity in ΔOD calculation and curve fitting was that two data recording lengths were chosen for each interrogated location on the crystal, 0.05 sec. and
2.0 sec. The most accurate ΔOD calculations came from the short time data runs, because the long ones had data points that were spaced further apart in time and thus the oscilloscope could potentially have missed the highest voltage data points. Accordingly, all reported ΔOD values were calculated using the 0.05 sec. data recordings. All curve fitting was performed using the 2.0 sec. data recordings except for 785 nm where a 5.0 sec. data recording length was necessary to see the steady state.

Fitting decaying exponentials to experimental data revealed the relative amount and types of electrons and holes participating in grating formation. Multiple areas in the crystal were interrogated at each wavelength, but for brevity and uniformity only area 2 will be closely examined and discussed in this paper. Curve fitting was performed in both MATLAB and Origin and as they agree quite closely with one another, only the Origin curve fitting parameters are presented and used for determining charge carrier participation percentages and lifetimes. Self-pumped counter-propagating two-beam coupling measurements were acquired for at least two power levels for each wavelength. Several different data acquisition methods were attempted before an accurate method was determined, which holds the number of photoexcited charge carriers approximately equal amongst all wavelengths. This is important, because it means the relative intensities, ΔODs, and lifetimes are in a comparable regime, allowing us to make valid and accurate comparisons.

For the first data set, each laser was attenuated to 5mW to obtain a baseline. For the second set, the power levels were roughly adjusted relative to 532 nm at 5mW as shown in Table 4.1 in an early and inaccurate attempt to take into account the effect of the absorption coefficient.
Table 4.1: Absorption coefficients & power levels for method 2

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Absorption coefficient (cm⁻¹)</th>
<th>Power (mW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>447</td>
<td>3.22715</td>
<td>8.43</td>
</tr>
<tr>
<td>523</td>
<td>2.00354</td>
<td>5.24</td>
</tr>
<tr>
<td>532</td>
<td>1.91312</td>
<td>5.0</td>
</tr>
<tr>
<td>561</td>
<td>1.60495</td>
<td>4.19</td>
</tr>
<tr>
<td>633</td>
<td>0.82136</td>
<td>2.15</td>
</tr>
<tr>
<td>785</td>
<td>0.16834</td>
<td>0.44</td>
</tr>
</tbody>
</table>

The third method of data collection assumed that virtually all absorption originates from the Fe²⁺ and Fe³⁺ dopant atoms and that this was the only important factor. Data was acquired for a select number of wavelengths with each power level adjusted relative to the absorption coefficient of 532 nm with the desired intent to excite approximately the same number of charge carriers at each wavelength. 532 nm was again anchored at 5mW and all the other lasers were adjusted as indicated in Table 4.2.

Table 4.2: Absorption coefficients & power levels to excite the same number of charge carriers at each wavelength

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Absorption coefficient (cm⁻¹)</th>
<th>Power (mW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>447</td>
<td>3.22715</td>
<td>2.96</td>
</tr>
<tr>
<td>523</td>
<td>2.00354</td>
<td>4.77</td>
</tr>
<tr>
<td>532</td>
<td>1.91312</td>
<td>5.0</td>
</tr>
<tr>
<td>561 (NOT OBTAINED)</td>
<td>1.60495 (NOT OBTAINED)</td>
<td>5.96</td>
</tr>
<tr>
<td>633</td>
<td>0.82136</td>
<td>11.65</td>
</tr>
<tr>
<td>785 (NOT OBTAINED)</td>
<td>0.16834 (NOT OBTAINED)</td>
<td>56.82</td>
</tr>
</tbody>
</table>

The final and most accurate method, was to acquire data with each power level adjusted to hold the relative intensities inside the crystal equal so as to excite approximately the same number of charge carriers at each wavelength. The powers were adjusted according to, \( \frac{P_1 \lambda_1 \alpha_1}{w_1^2} = \frac{P_2 \lambda_2 \alpha_2}{w_2^2} \), where \( P \) is power, \( \lambda \) is wavelength, \( \alpha \) is the absorption coefficient, and \( W \) is the focused beam waist radius\(^{24} \). 532 nm was anchored at 5mW and all the other lasers were adjusted as indicated in Table 4.3. To find the ideal power level for another wavelength simply solve for \( P_1 = \frac{P_2 \lambda_2 \alpha_2 W_1^2}{W_2^2 \lambda_1 \alpha_1} \) where everything
with the subscript 1 relates to the new wavelength and everything with the subscript 2 relates to 532 nm. The data obtained from the “Actual power” column was acquired before the ideal power formula was discovered, but was so close to the ideal power that reacquisition was not necessary. Data for 561 and 785 nm was not acquired using this method and thus no data for them is included in Table 4.3.

Table 4.3: Important laser variables for accurately determining power levels in order to excite equal numbers of charge carriers

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>447</th>
<th>523</th>
<th>532</th>
<th>561</th>
<th>633</th>
<th>785</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waist radius after lens (µm)</td>
<td>41.36</td>
<td>39.93</td>
<td>35.39</td>
<td>256.02</td>
<td>56.81</td>
<td>72.68</td>
</tr>
<tr>
<td>Absorption coef. (cm⁻¹)</td>
<td>3.22715</td>
<td>2.00354</td>
<td>1.91312</td>
<td>1.60495</td>
<td>0.82136</td>
<td>0.16834</td>
</tr>
<tr>
<td>Ideal Power (mW)</td>
<td>4.8</td>
<td>6.2</td>
<td>5</td>
<td>73.9</td>
<td>25.2</td>
<td>40.6</td>
</tr>
<tr>
<td>Actual Power (mW)</td>
<td>5</td>
<td>5.2</td>
<td>5</td>
<td>5</td>
<td>24.4</td>
<td>5</td>
</tr>
<tr>
<td>Ratio of $P_{\text{Ideal}}/P_{\text{Real}}$</td>
<td>0.96</td>
<td>1.192308</td>
<td>1</td>
<td>14.78</td>
<td>1.032787</td>
<td>8.12</td>
</tr>
<tr>
<td>Theoretical ΔOD with single charge carrier</td>
<td>2.20</td>
<td>2.08</td>
<td>2.04</td>
<td>2.20</td>
<td>1.95</td>
<td>1.33</td>
</tr>
<tr>
<td>Actual ΔOD</td>
<td>1.6-1.9</td>
<td>1.5-1.6</td>
<td>1.9-2.2</td>
<td>1.4-1.7</td>
<td>1.5-1.6</td>
<td>0.6-0.7</td>
</tr>
</tbody>
</table>

Absorption spectra for the VLOC K83B2-3 crystal was acquired on a Cary 5000 UV-Vis-NIR Spectrometer for many locations polarized along the a-axis to verify that the data presented below is representative of the crystal as a whole in the a-c plane.

Figure 4.1: Absorption spectra for VLOC K83B2-3 KNbO₃:Fe crystal
One potential source of error in using the absorption coefficient to adjust power levels is found in following the approach of reference 25, wherein the authors saw that the maximum energy exchange. This implies the location of the largest portion of the space-charge field was occurring close to the front face of the crystal. Additionally, this maximum space-charge field location appears to be affected by the incident pump power and wavelength, indicating that the wavelength and power level alter the gain medium length in the crystal for signal and pump interaction. As a result, the incoming photons see different numbers of donor and trap densities depending on their energy (i.e. electron-volts) and their overall numbers (i.e. laser power level). This means the absorption spectra obtained in the Cary 5000 was slightly different from the absorption spectra seen in the PR two beam coupling experiment, because the Cary 5000 data is accurate for low power light passing through the entire crystal. Thus, our use of absorption spectra data to calculate adjustments in the lasers’ power levels in attempting to excite an equal number of charge carriers was not entirely accurate. Nevertheless, the method used in this thesis follows the general principles needed to meet this requirement and is likely very close to the actual power levels.

The plots and curve fitting parameters for all wavelengths are shown below. Curve fitting has been performed using the decaying exponential formula,

\[ y = A_0 + A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2} + A_3 e^{-t/\tau_3}. \]  

(4.1-1)

Multiple curve fitting parameters are included from data runs performed on different days all within 0.5 mm of the same spot on the crystal. As our purpose is to determine how the different charge carriers are contributing to the creation of the net space charge field at different wavelengths, we have performed curve fitting with the minimum and
maximum number of possible decaying exponential terms as illustrated in Figure 1.1; the fitting parameters are in Table 4.4 – 4.7. PR two-beam coupling is shown Figure 4.2 – 4.3. In both sets of figures we can clearly see the effects of the various absorption coefficients on the transmitted power. 447 nm ($\alpha = 3.23$ cm$^{-1}$) has a relatively large absorption coefficient compared to the other wavelengths and as such, much less light is transmitted due to linear absorption.

Figure 4.2: Experimental data plots of 5 mW baseline. Exploded view on the right.
Table 4.4: Baseline curve fitting parameters. Minimal number of terms used for curve fitting.

<table>
<thead>
<tr>
<th>Power (mW)</th>
<th>Wavelength (nm)</th>
<th>Lifetimes (millsec.)</th>
<th>Coefficient</th>
<th>AOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>447</td>
<td>1.44 ( \tau_1 ) 11.80 ( \tau_2 )</td>
<td>0.01513 A1 0.00802 A2</td>
<td>1.5534</td>
</tr>
<tr>
<td></td>
<td>2.774 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>523</td>
<td>1.27 ( \tau_1 ) 16.22 ( \tau_2 )</td>
<td>0.07769 A1 0.02898 A2</td>
<td>1.2974</td>
</tr>
<tr>
<td></td>
<td>2.371 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.60 ( \tau_1 )</td>
<td>0.11112 A1 1.8546</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>1.63 ( \tau_1 )</td>
<td>0.09704 A1 1.8947</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.63 ( \tau_1 )</td>
<td>0.10376 A1 2.0779</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>561</td>
<td>6.04 ( \tau_1 )</td>
<td>0.15587 A1 1.4648</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.210 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>633</td>
<td>4.03 ( \tau_1 )</td>
<td>0.33179 A1 1.3051</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.959 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>785</td>
<td>37.69 ( \tau_1 )</td>
<td>0.49005 A1 0.5779</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.580 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 4.5: Baseline curve fitting parameters. Maximum number of terms used for curve fitting

<table>
<thead>
<tr>
<th>Power (mW)</th>
<th>Wavelength (nm)</th>
<th>Lifetimes (millsec.)</th>
<th>Coefficient</th>
<th>AOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>447</td>
<td>1.3 ( \tau_1 ) 9.64 ( \tau_2 ) 173.9 ( \tau_3 )</td>
<td>0.01435 A1 0.00867 A2 0.000785 A3</td>
<td>1.5534</td>
</tr>
<tr>
<td></td>
<td>2.774 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>523</td>
<td>0.84 ( \tau_1 ) 2.32 ( \tau_2 ) 18.37 ( \tau_3 )</td>
<td>0.05383 A1 0.03198 A2 0.02616 A3</td>
<td>1.2974</td>
</tr>
<tr>
<td></td>
<td>2.371 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.64 ( \tau_1 ) 3.31 ( \tau_2 )</td>
<td>0.07103 A1 0.03706 A2</td>
<td>1.8546</td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.57 ( \tau_1 ) 3.40 ( \tau_2 )</td>
<td>0.08503 A1 0.03964 A2</td>
<td>1.8947</td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.33 ( \tau_1 ) 3.17 ( \tau_2 )</td>
<td>0.09722 A1 0.01986 A2</td>
<td>2.0779</td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.31 ( \tau_1 ) 2.71 ( \tau_2 )</td>
<td>0.12029 A1 0.01985 A2</td>
<td>2.1653</td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>561</td>
<td>3.31 ( \tau_1 ) 27.76 ( \tau_2 )</td>
<td>0.14876 A1 0.03547 A2</td>
<td>1.4648</td>
</tr>
<tr>
<td></td>
<td>2.210 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>633</td>
<td>1.83 ( \tau_1 ) 13.35 ( \tau_2 )</td>
<td>0.31435 A1 0.10076 A2</td>
<td>1.3051</td>
</tr>
<tr>
<td></td>
<td>1.959 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>785</td>
<td>24.47 ( \tau_1 ) 81.73 ( \tau_2 )</td>
<td>0.34890 A1 0.18223 A2</td>
<td>0.5779</td>
</tr>
<tr>
<td></td>
<td>1.580 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 4.3: Experimental data plots where excited charge carrier numbers are held constant. Exploded view on the right.

Table 4.6: Charge carrier numbers equal curve fitting parameters. Minimal number of terms used for curve fitting.

<table>
<thead>
<tr>
<th>Power (mW)</th>
<th>Wavelength (nm)</th>
<th>Lifetimes (millsec.)</th>
<th>Coefficient</th>
<th>AOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>447</td>
<td>1.44 τ1</td>
<td>0.01513 A1</td>
<td>1.5534</td>
</tr>
<tr>
<td></td>
<td>2.774 eV</td>
<td>11.80 τ2</td>
<td>0.00802 A2</td>
<td></td>
</tr>
<tr>
<td>5.24</td>
<td>523</td>
<td>1.10 τ1</td>
<td>0.07522 A1</td>
<td>1.597</td>
</tr>
<tr>
<td></td>
<td>2.371 eV</td>
<td>12.29 τ2</td>
<td>0.03200 A2</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.60 τ1</td>
<td>0.11112 A1</td>
<td>1.8546</td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>1.63 τ1</td>
<td>0.09704 A1</td>
<td>1.8947</td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.63 τ1</td>
<td>0.10376 A1</td>
<td>2.0779</td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.60 τ1</td>
<td>0.11112 A1</td>
<td>2.1653</td>
</tr>
<tr>
<td></td>
<td>2.331 eV</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>24.4</td>
<td>633</td>
<td>1.34 τ1</td>
<td>1.38581 A1</td>
<td>1.5044</td>
</tr>
</tbody>
</table>
Table 4.7: Charge carrier numbers equal curve fitting parameters. Maximum number of terms used for curve fitting

<table>
<thead>
<tr>
<th>Power (mW)</th>
<th>Wavelength (nm)</th>
<th>Lifetimes (millsec.)</th>
<th>Coefficient</th>
<th>ΔOD</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>447</td>
<td>1.3 τ1 9.64 τ2 173.9 τ3</td>
<td>0.01435 A1 0.00867 A2 0.000785 A3</td>
<td>1.5534</td>
</tr>
<tr>
<td>5.24</td>
<td>523</td>
<td>0.63 τ1 1.44 τ2 12.81 τ3</td>
<td>0.02951 A1 0.04999 A2 0.03059 A3</td>
<td>1.597</td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.64 τ1 3.31 τ2</td>
<td>0.07103 A1 0.03706 A2</td>
<td>1.8546</td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.57 τ1 3.40 τ2</td>
<td>0.08503 A1 0.03964 A2</td>
<td>1.8947</td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.33 τ1 3.17 τ2</td>
<td>0.09722 A1 0.01986 A2</td>
<td>2.0779</td>
</tr>
<tr>
<td>5</td>
<td>532</td>
<td>0.31 τ1 2.71 τ2</td>
<td>0.12029 A1 0.01985 A2</td>
<td>2.1653</td>
</tr>
<tr>
<td>24.4</td>
<td>633</td>
<td>0.56 τ1 7.24 τ2</td>
<td>1.55850 A1 0.30552 A2</td>
<td>1.5044</td>
</tr>
</tbody>
</table>

4.2 Interpretation of Results

In the data above we see several interesting findings. First, ΔOD for 532 nm (2.33 eV) is consistently higher than for any of the other wavelengths throughout the visible spectrum despite the fact that the absorption at 532 nm is less than that at 447 nm (2.77 eV). One possible explanation for this can be seen in Figure 1.1. At 2.33 eV we may not yet be creating any holes to compete with the electron formed grating, whereas at 523 nm (2.37 eV) and 447 nm we almost certainly have hole competition. At 2.33 eV we are using the maximum amount of energy possible to excite electrons at E₁=0.85 eV, while exciting few to no competing holes at E₂=2.35 eV. Also, with incoming photons at 2.33 eV, the E₁ excited electrons (2.33 eV – 0.85 eV) arrive at the conduction band (CB) with 1.48 eV of additional energy to move them higher into the CB. The photoexcited electrons in the CB are energetic and moving very fast, which benefits photorefractive grating formation. Additionally, a previously well-established computational model,
which only includes a single charge carrier for grating formation predicts a ΔOD of 2.1 for 447 nm, but experimentally the ΔOD ranges from 1.6 to 1.9. The source of this discrepancy is likely because the old model does not take into account the competing holes created at 2.35 eV, which counteract the grating formed by the electrons that have activation energies of 0.85 eV and 2.54 eV.

We expected to use only one decaying exponential term to fit the data at lower energies, but were surprised to find that two terms were required to make a good visual curve fit. This could indicate that at the lower photon energies an electron “retrapping” model may be appropriate. This can be understood in that some portion of the electrons photoexcited to the CB do not have enough velocity/energy along a vector towards the dark fringes to reach them before becoming retrapped in the bright fringes. This implies that a higher concentration of Fe$^{3+}$ traps inside the crystal could cause more retrapping to occur.$^{26}$ After being retrapped, they are then again photoexcited to the CB and complete their journey to the dark fringes. “Hotter” or more energetic electrons in the CB would be much more likely to make it all the way to the dark fringes without being retrapped. Somewhat similar to observing how a fast moving ball on a board with holes in it does not fall through the holes, but a slow moving ball does. For example, 1.96 eV photons (633 nm at 24.4 mW) only require 0.85 eV to excite electrons from the E$_1$ Fermi band to the CB and with the remaining 1.11 eV pushing the electrons higher into the CB.

According to the retrapping model this may not be enough energy to get all the electrons to the dark fringes before being retrapped. In the simple decaying exponential model, the $A_2$ term in the Origin curve fitting parameters of Table 4.7 for 633 nm would likely be related to the retrapped electrons. Retrapping may be modeled by a variety of techniques
and methods. Huber et al. in reference 26, used stretched exponentials to model relaxation rates, which are analogous to retrapping and in future work should be examined in great detail. The retrapping model would also mean that a significant number of electrons at 2.77 eV (447 nm) would be retrapped, because the electrons with activation energy $E_3=2.54$ eV would absorb almost all the energy just to reach the CB, leaving only 0.23 eV to push the electrons deeper into the CB.

Another possible explanation could be that collisional diffusion at the longer wavelengths (532, 561, 633, and 785 nm) homogeneously broadens out the range of electron velocity vectors in such a way that the Cauchy-Lorentz (C-L) model is more appropriate to fit the data. C-L is usually appropriate when homogeneous natural broadening as a result of finite radiative lifetime is present and/or collisional broadening is present. The C-L probability distribution function (PDF) is 

$$\frac{1}{\tau} = \frac{w}{\pi[(\tau-\tau_0)^2+w^2]}.$$  

Where $w$ is the half width at half maximum (HWHM) and $\tau_0$ specifies the peak of the distribution. Simply insert the PDF into $e^{-t/\tau}$ to obtain $e^{-t/\tau}w/(\pi[(\tau-\tau_0)^2+w^2])$. Collisional diffusion is of course occurring at all wavelengths, but at the energies of 1.96 eV and 2.21 eV and to a lesser degree 1.58 eV, which are all far from the hole excitation level, the simple single term curve fit using $e^{-t/\tau}$ simply breaks down, while a single term C-L term fits very nicely. Even at 2.33 eV, which is close to the hole excitation level of 2.35, a single term C-L fits the data much better than a simple decaying exponential. It must also be noted that single term C-L curve fitting does not fit the data well at 2.37 eV nor at 2.77 eV. This is most likely because of the additional charge carrier species involved in grating formation. Quite possibly two or three term C-L curve fitting would fit these higher energies, but this is beyond the scope of this thesis and will be left for future work.
Attempts to modify Origin’s curve fitting code to match the C-L probability distribution function were very promising, but are still limited. Future improvements in understanding the C-L PDF and applying it to modeling techniques may yield more definitive results and provide insightful and accurate quantitative data on carrier lifetimes. A comparison of curve fittings laid over data plots using both simple exponential and C-L techniques is seen in Figure 4.4 – 4.5. The adjusted coefficient of determination, $R^2$, which indicates how well the curve fitting matches the actual data plots is included in the figures. An $R^2$ value of 1.0 would indicate a perfect fit.

Figure 4.4 Single term curve fitting. A simple decaying exponential was used on the left and Cauchy-Lorentz on the right. Continued on the next page.
Another finding was that in the grating formation data plots of 447 nm and 523 nm we expected to see a steep fall in transmission power to an initial low value followed by a gradual rise in transmission power as the holes formed at ~2.35 eV begin to recombine with electrons in the dark fringes. The expected delay in rising to a steady state would come from the fact that hole mobility is far less than that of electron mobility and it is strongly believed that the holes play a detrimental role in grating formation of reduced KNbO₃:Fe. However, we did not observe any delayed rise in the transmission power for any wavelength and this may be explained by the fact that in the three term curve fit equation (3.1-1) for 447 nm (5 mW), \( \tau_3=173.9 \text{ ms} \) is much larger than \( \tau_1=1.3 \text{ ms} \), or \( \tau_2=9.64 \text{ ms} \) and \( A_3=0.000785 \) is much less than \( A_1=0.01453 \), or \( A_2=0.00867 \). Below are several computer generated plots where \( \tau_3 \) and \( A_3 \) were given opposite signs and allowed to vary, which clearly demonstrate the relationship between the delayed rise to steady-state and these variables. For the computer generated plots in Figure 4.6, real
Origin generated two-term curve fitting parameters for 447nm at 5 mW were used with an additional third term decaying exponential added in to represent the holes.

In the case of holes in the valence band, we likely have a combination of a small coefficient (i.e. not many holes participating in comparison to the electrons) and a long lifetime (i.e. slow mobility when compared to the electrons in the CB). It is also important to note that the band gap of KNbO$_3$ is indirect and crystal inhomogeneity may affect energy level transitions. Ambient room temperature energy will shift the energy on the valence band by $k=0.1288 \text{ }\mu\text{m}^{-1}$ (at Temp=295 K, 48.7783 $\mu\text{m}$ or 0.0254 eV) to the right. An exhaustive literature search did not yield any band gap diagrams of KNbO$_3$:Fe, so we do not know exactly how many eV the system will be lowered by. This may be important, because 447 nm (2.77 eV) is off from the 2.35eV valence band charge carrier center by 0.42 eV and unless the room temperature energy can shift the crystal gap down by 0.42 eV, which seems unlikely, we may not create a large enough number of holes to see their affects in the plots, although we certainly do see that the holes lower the overall $\Delta\text{OD}$ at 447 nm.
In relation to the graphs in Figure 4.6, this means that though we have holes being generated at 447 nm their number may be few, causing our experimental data plots in Figure 4.7 to dip down very little, potentially being lost in system noise. Even if there is a slight dip it would rise so slowly that it may be impossible to view in the lifetime of the oscilloscope data capture window. Such a rise is also unseen in the experimental data plots for 523 nm and 532 nm. We believe that the noise seen in Figure 4.8 in the steady state of the 523 nm plot was caused by electro-magnetic interference from an unknown source around our laboratory.

Figure 4.7: 447 nm data plots with two and three term curve fitting performed in the left and right images respectively.

Figure 4.8: Data plots overlaid with curve fitting for maximum possible charge carriers at 523 nm and 532 nm. No evidence of hole competition is visible.
We have attempted to fit two term decaying exponentials to the experimental data of 447 nm down to ~1% of difference between peak transmission voltage and steady state voltage. Those curve fitting parameters/equations were then used to extrapolate what a plot of those parameters would look like graphed all the way out to the end of the data window as shown in the middle image of Figure 4.9. Afterwards, in the right image of Figure 4.9, we subtracted that extrapolation from the real data in the hope that we might see some slight dip or rise indicating the presence of competing detrimental holes. Nothing indicating their presence has been observed in any of the 447 nm power levels, except for 3 mW where a very slight bump is seen. It is unlikely that this bump can be attributed to holes, which should have a long-term rising effect in the plot and not merely a small bump as seen in the plot.

*Figure 4.9: Left figure is the original data. Middle figure is the extrapolated curve. Right figure is original-extrapolation*
CHAPTER 5
CONCLUSION & FUTURE WORK

We have examined in detail the multiple charge carrier centers in KNbO$_3$:Fe involved in photorefractive grating formation through self-pumped counter-propagating two-beam coupling measurements at multiple wavelengths and power levels. We have shown that physical interactions of charge carriers in two-beam coupling at longer wavelengths is intrinsically different than the physical interactions at shorter wavelengths and have proposed several theories and models for what may be occurring. Clearly, the single species of charge carrier involved in grating formation at 532, 561, and 633 nm cannot be modeled with a simple single decaying exponential $e^{-t/\tau}$ and one must turn to the retrapping model, or some form of homogeneous velocity broadening that requires the Cauchy-Lorentz model. Our methods also revealed that 532 nm is particularly effective at creating fast gratings with high $\Delta$ODs. These findings provide a few steps towards understanding and tailoring KNbO$_3$ crystals for improved and predictable PR responsivity.

Future work might focus on fully developing a computer model with multiple charge carrier species, which accurately simulates the growth of PR gratings. It may prove beneficial to look more deeply at the space-charge field gradient depths of the individual wavelengths and power levels to more accurately excite equal numbers of
charge carriers. Additionally, we hope improvements will be made in the Cauchy-Lorentz model to obtain better curve fitting results and in connection with this, to examine avalanche photodiode type electron pair production. As high energy fast moving electrons in the CB move towards the dark fringes they may collide with lower energy electrons in the Fermi-level and excite them into the CB. Finally, thermodynamic aspects of the retrapping model should be studied in more detail. A cursory thermodynamic analysis can be found in Appendix D.
REFERENCES


APPENDIX A

MATHEMATICA CODE

Program to Calculate the Effective EO, Gain Coefficients, and ΔOD in Fe:KNbO3
with Self-Pumped Counter-Propagating Two-Beam Coupling

(Original Coding in MathCAD by Gary Cook as found in Gary’s old binder marked “MathCAD 3” entitled
“Test fit for off-axis Fe:KNbO3 paper to determine the values of gamma and Idark along the c-axis.”
Rewritten in Mathematica by Ighodalo Idehenre. Final version Modified and expanded by Michael
Amonson. Special thanks to W. Joshua Kennedy for coding assistance.)

ClearGlobal[] := (ClearAll["Global`*"] ; Clear[Derivative] ; );
RemoveGlobal[] := (ClearGlobal[] ; Remove["Global`*"] ; );

(*Physical Constants;
c Speed of Light*;
e Charge of an Electron;
h Planck’s Constant;
Subscript[\(\epsilon\), 0] Permittivity Of Free Space;
Subscript[\(\phi\), B] Boltzmann's Constant; *)
\(c=2.99792458\times10^8;\text{(*Meters/Second*)}\)
\(e=1.60217662\times10^{-19};\text{(*Coulombs*)}\)
\(h=6.62607004\times10^{-34};\text{(*Joules*Seconds*)}\)
\(\epsilon_0=8.85418782\times10^{-12};\text{(*Farads/Meter*)}\)
\(k_B=1.38064852\times10^{-23};\text{(*Joules/Kelvin*)}\)

(*User Defined Variables;*

Subscript\([w, o]\) Pump Beam Spot Size Radius Before Lens;

\(f\) Lens Focal Length;

Subscript\([P, in]\) Input Power of Pump (i.e. Power output of the laser);

\(\text{reff}\) Effective Electro-Optic Coefficient;

Subscript\([\gamma, r]\) Recombination Coefficient;

Subscript\([\tau, di]\) Dielectric Relaxation Time;

\(\mu\) Carrier Mobility;

\(a\) Distance Between Lens And Crystal Front Surface;

\(k\) Propagation Constant In Medium;

\(n\) Refractive Index;

\(R_f\) Front Face Reflectance;

\(R_b\) Back Face Reflectance;

Subscript\([F, number]\) F Number;

Subscript\([w, o]\) Pump Beam Spot Size Radius Before Lens;

Subscript\([w, op]\) Pump Beam Spot Size After Lens;

\(\sigma\) Absorption Cross Section Per Unit Energy;

Subscript\([I, Dark]\) Dark Current Expressed In Terms Of Intensity;

\(L\) Length of the Crystal;

\(ND\) Total Fe number density(Fe\(^{2+}\) and Fe\(^{3+}\));

\(NA\) Acceptor(Fe\(^{3+}\)) number density. Trap density used to control the saturation field;

Subscript\([\epsilon, rs]\) Relative Static Permittivity of Subscript\([\text{KNbO}, 3]\);

\(\epsilon_{\text{eff}}\) Effective Dielectric Permittivity Constant of Subscript\([\text{KNbO}, 3]\);

\(\Lambda\) Grating Spacing;

\(\nu\) Frequency;

\(z_0\) Position of Pump Beam at Front of Crystal;

\(z_L\) Position of Pump Beam at End of Crystal;

\(z_{\text{Rp}}\) Rayleigh Range After Lens;

\(W_0=\left(3.3/2\right)\times10^{-3};\text{(* Beam Waist Radius in Meters*)}\)

\(f=200\times10^{-3};\text{(*Meters*)}\)

\(P_{\text{in}}=5\times10^{-3};\text{(*Power output of laser in Watts*)}\) (*CHECK THIS VALUE*)

\(r_{13}=28\times10^{-12};\text{(*Meter/Volt. This value is for pure KNbO3 & was obtained from the CRC Handbook of Chemistry & Physics, 97 Edition, pg.12-174*)}\)

(* r_{13} might be different when KNbO3 is doped with 2000ppm in the melt *)

\(\text{reff}=r_{13};\text{(* reff for light polarized in the A-C plane entering parallel to the A-C plane at normal incidence simplifies to r_{13} *)}\)

\(T=295;\text{(*Kelvins*)}\)

\(\lambda=785\times10^{-9};\text{(*Meters*)}\)
a=(f-0.012)-L/3; (*Meters. The Back Focal Length is 0.188m, but the Effective Focal Length is 0.200m*)
k=(2*π*n)/λ; (*1/Meters*)
nc=2.3813; (*Refractive index of crystal's c-axis*)
na=1.00029; (*Refractive index of air at STP (i.e. standard temp=273.15K=0°C, pressure=1atm=101.325kPa]*)
n=nc;
Rf=((nc-na)/(nc+na))^2;
Rb=((na-nc)/(na+nc))^2;
Fnumber=f/(2*W0); (*Dimensionless*)
Wop=(f*λ)/(π*W0); (*Meters*)
Iin=Pin/(π*(Wop)^2); (*Intensity after the Gaussian beam has passed through the lens. Watts/Meter^2*)
θ=0; (*The incoming laser beam is normal to the crystal face*)
σ=8.6*10^-4; (*Meters^2/Joule*) (*According to Sergey, IMPOSSIBLE TO KNOW THIS WITH ANY DEGREE OF CERTAINTY*)
IDark=10000; (*Watts/Meters^2*) (*CHECK THIS VALUE. It is a fudge factor*)
Id=IDark;
L=5.25*10^-3; (*Meters*) (*DIFFERENT FOR EACH CRYSTAL*)
ND=5*10^17; (*CHECK THIS VALUE. The dopant level affects this*)
NA=1.1*10^23; (*Carrier/Meter^3*)
(*) NA[ND_,α_,ν_,σ_]:=ND-α/(h*ν*σ); Carrier/Meter^3. This is only valid if ND is donor density, not total Fe2+ & Fe3+ density.*)
ε_{eff}=73-46*Cos[2*θ]+19*Cos[4*θ]-12*Cos[6*θ]+3*Cos[8*θ]-2*Cos[10*θ]
ε_{s}=ε_{eff}/ε_{0}; (*Farads/Meter*)
ν=c/λ; (*Hertz. The frequency needs to be tied to α, which is also tied to σ the absorption cross section*)
Λ=λ/(2*n); (*Meters*)
Λ_{opt}=2*π*W_{0}; (*Meters*)
Γ=5000;
N_{trapDensity}=Γ*l*ε_{s}/(n^3*ε_{eff}*e*Λ);
z0=a;
zL=a+L;
zR=(π*Wop^2)/λ; (*Meters*)
I_{0}=Pin/(π*(2.34/2)^2/1000000) (*Intensity before the beam is focused by the lens. Watts/Meter^2*)
I_{0}=Pin/(π*(0.0579/2)^2/1000000) (*Intensity after the beam is focused by the lens. Watts/Meter^2*)
1162.65
1.89899*10^6
N[Λ]
\((\pi \times 0.225^2)\)
\[1.64826 \times 10^{-7}\]
0.159043


E0\[\square\] Externally Applied Electric Field;
ED\[\square\] Diffusion Field;
EPV\[\square\] Photo Voltaic Field;
ED\[\square\] Diffusion Field. Electrons excited into the conduction band naturally move from the bright fringe areas where they were created and are in great abundance to the dark fringe areas where no electrons are being excited into the conduction band;
Subscript[E, M]\[\square\] Drift Field. Related to the externally applied Electric field. Movement of charge carriers, which is due to an applied electric field, often stated as the electromotive force over a given distance. When an electric field is applied across a semiconductor material, a current is produced due to the flow of charge carriers;
EQ\[\square\] Saturation Field. Maximum applied electric field beyond which electron diffusion velocity no longer increases.;
Subscript[E, S]\[\square\] Space Charge Field;
Λ\[\square\] Grating spacing;
NA\[\square\] Trap density used to control the saturation field;
ND\[\square\] Total Fe Donor Concentration;
Γ\[\square\] Gain Coefficient/Intrinsic Coupling Coefficient;
τ\[\square\] Dielectric Relaxation Time/Response Time. Depends and varies on local values of beam intensity;
Subscript[γ, r]\[\square\] Charge Recombination Coefficient;
E0\[\square\] Externally Applied Electric Field;
EPV\[\square\] Photo Voltaic Field;
σ\[\square\] Absorption Cross Section Per Unit Energy;
α\[\square\] Linear Absorption Coefficient; (*)

\(E_0^{incr}=0;\)
\(E_0=\text{Table}[0,\{E_0^{incr}+1\}];\)
(* This is when \(E_0\) is allowed to vary \(\square\) Table[Eomin+(Eomax-Eomin)/Eoincr*ii,\{ii,0,Eoincr\}]\*10^5; *)

(* In my case there is no external field, but \(\alpha\) should vary here with different wavelengths which would also cause NA to vary. NA then in turn causes Subscript[E, M], EQ, Subscript[E, S], Γ, and \(\tau\) to vary. I want to plot ΔOD vs wavelength or frequency. What I need to determine is how \(\alpha\) varies with wavelength. I can learn this from the Cary 5000. But what I really want to know is what percentage of each charge carrier is participating at a given wavelength and that wavelength's associated \(\alpha\). *)

EPV=0;(*Volts/Meter*) (*CHECK THIS VALUE*) (*Photo Voltaic electric field for KNbO3 is negligible*)

\(ED[T,Λ]=:(2\pi\times k_B\times T)/(e \times Λ);(*Volts/Meter*)\)
\[ EM[k, NA, \mu, \gamma_r] := \gamma_r NA / (2\mu k) ; (* Volts/Meter *) \]
\[ EQ[\Lambda, NA, ND, \varepsilon_s] := (\Lambda \varepsilon_s NA) / (2\pi \varepsilon_s) ; (* Volts/Meter *) \]
\[ ESc[E_0, ED, EPV, EQ, NA, ND] := (E_0 + iED + EPV) / (1 + ED/EQ - I*(E_0/EQ + NA/ND*EPV/EQ)) ; (* Volts/Meter *) \]

(* Subscript[E, sc] simplifies to i*ED/(1+ED/EQ) --- G. Cook, High Optical Gain Using Counterpropagating... *)
\[ \Gamma[ES, k, n, reff] := k * n^2 \text{reff} \text{Im}[ES] ; \]
\[ \tau[ED, EM, E_0, EPV, EQ, NA, ND, \tau_{di}] := \tau_{di} \text{Re}[(1 + ED/EM + iE_0/EM) / (1 + ED/EQ - I*(E_0/EQ + NA/ND*EPV/EQ))] ; (* Seconds *) \]

**Steady State \( \Delta OD \) Calculations**

\[ I_{\text{pump, in}} = \frac{P_n}{\pi (w_{cp})^2} \]

\[ \text{Photorefractive Window} \]

\[ I_{\text{signal, out}} = ? \]
\[ I_{\text{signal, 0}} = ? \]
\[ I_{\text{signal, in}} = 0 \]
\[ I_{\text{pump, 0}} = ? \]
\[ I_{\text{pump, out}} = ? \]

(* While the boundary conditions (intensities) vary during the build up of the grating, eventually the system reaches a steady state. At this point these values should remain static resulting in a spatial boundary value problem. In this application we know the pump (incoming beam) intensity at the front face/boundary but not the signal (beam reflected off of the back face of the crystal). Simultaneously, we know the signal intensity at the back face/boundary but not the pump. The possible values of both unknowns are however constrained by the input. Under paraxial conditions (i.e. a plane wave) the maximum power/intensity of the pump beam inside the front face of the crystal cannot exceed that of the transmitted pump ((1 - Subscript[R, f])*I_{in}). Likewise, the minimum power/intensity of the signal beam at the front face of the crystal cannot be lower than the transmitted pump after it has been reflected off the back surface(Rb), attenuated by the medium \( e^{2\alpha(zL-z0)} \), and transmitted through front surface((1 - Subscript[R, f])*I). Using an average of these two limits, we can guess the intensity of the signal at the front boundary \( I_{sig0} \) allowing us to address the boundary value problem (BVP) as an initial value problem (IVP) and solve it using IVP techniques.

Since \( I_{sig0} \) (signal just inside the front face of the crystal) was a guess, the calculated signal power/intensity at the back surface (Subscript[I, sigL]) will probably not be equal to the calculated pump power/intensity reflected by the back surface(I_{pumpL}*Rb). If the difference is negative, it means we guessed too high. We can then define the upper limit of the signal to be our initial guess. If the difference between Subscript[I, sigL] and I_{pumpL}*Rb is positive, it means our guess was too low, and we change the lower limit to our initial guess. By repeating this process we can eventually converge on the solution to the BVP. This is what is referred to as the shoot and match method. *)
(*Routine for various values of EPV*)

freq=Table[i,{i,447,785,1.086}] (*freq=Table[i,{i,447,785,1.086}]*)
α=Table[i,{i,3.28,0.168,-0.01}] (*1/Meters*)

freq=Table[i,{i,785,786,1}]
α=Table[i,{i,0.16834,0.16835,0.00001}] (*1/Meters*)
Transpose[{freq,α}]
{785,786}
{0.16834,0.16835}
{{785,0.16834},{786,0.16835}}
Dimensions[freq]
Dimensions[α]
{2}
{2}

ΔODfromAlpha={}; (* Creates an empty list for ΔOD values that will come from the "For" and "While" loops as we vary the absorption coefficient α *)

For[i=1,i<=Length[α],i++,
 (*Calculates Each output. Results in a 2-D matrix M[[ii,kk]]. The first argument (ii) corresponds to EO vector. For the second element, kk=1 returns the value EO[[ii]] while kk=2 returns the value of calculated parameter,i.e. Re{Subscript[E, s]} ,Im{Subscript[E, s]}, or Γ at E0[[ii]] *)
EScRe=Table[Re[ESc[E0[[ii]],ED[T,Λ],EPV,EQ[Λ,NA,ND,εs],NA,ND]],{ii,1,E0incr+1}];
EScIm=Table[Im[ESc[E0[[ii]],ED[T,Λ],EPV,EQ[Λ,NA,ND,εs],NA,ND]],{ii,1,E0incr+1}];
Γcal=Table[Γ[I*EScIm[[ii]]],{ii,1,E0incr+1}];
ΔODList=Table[0ii,{ii,1,E0incr+1}];

Do[
Ipump0=(1- Rf)*Iin;(*Pump after being transmitted through front.*)
Isig0max=Iin(Ipump0)*;(*The maximum possible value for signal is 100% reflection due to grating.*)
Isig0min=(1- Rf)Rb*E^{-2α[i]((zL-z0)*Ipump0};(*The minimum possible value for signal occurs when no grating exist.*)
Error=1;(*Metric for correcting the initial guess for the signal. Initially set to 1.*)
Acc=1*10^-5;(*Desired accuracy/difference between guess and calculated signal.*)

While[Abs[Error]>Acc,(*Loop will continue until accuracy threshold is met. If not use Alt+, to abort.*)
Isig0=(Isig0max+Isig0min)/2;
sols = (*NDSolve is mathematicas numerical IVP solver. This problem has two unknows and requires two initial values(Ip[z0],Is[z0]), and two differential equations(Ip'[z],Is'[z]) to solve.*)First[NDSolve[ {Ip[z] == -α[i]*Ip[z] - Γcal[[ii]] (Ip[z]IIs[z])/(Ip[z]+IIs[z]+IDark)-(2(z-f))'(zRp^2+(z-f)^2) Ip[z],IIs[z] == α[i]*IIs[z] -
\( \Gamma_{\text{cal}} \) 

\[
\frac{(Ip[z]*Is[z])/(Ip[z]+Is[z]+IDark) - (2(z-f))/(zRp^2+(z-f)^2)}{Is[z], Ip[z0] == Ipump0, Is[z0] == Isig0, \{Ip, Is\}, \{z, z0, zL\}}; \]

\( \text{IsigExp} = Rb*Ip[zL]/.sols; \) (*Signal at back surface should equal the backwards reflection of the calculated pump.*)

\( \text{IsigCalc} = Is[zL]/.sols; \) (*Actual signal inside the back face that was calculated by NDSolve.*)

\( \text{IpumpL} = Ip[zL]/.sols; \) (*Calculated Pump which is used for \( \Delta \text{OD} \]*)

\( \text{Error} = (\text{IsigExp} - \text{IsigCalc})/\text{IsigCalc}; \) (*Error between expected value and calculated*)

\[
\text{IpumpL} = \text{Ip}[zL]/.\text{sols}; \]

\( \text{If}[\text{Error} < 0, \text{Isig0max} = \text{Isig0}, \text{Isig0min} = \text{Isig0}]; \) (*Desides what the next guess should be.*)

\( \text{ΔODList}[[\text{ii}]] = \text{Log10}[(1-Rb)E^{-\alpha[[\text{ii}]]*(zL-z0)} \text{Ipump0}/(\text{IpumpL}(1-Rb))]; \) (*Calculates \( \Delta \text{OD} \]*)

\( \text{minimumpump} = \text{Ip}/.\text{sols[[1]]}[zL]; \) (* identifies the minimum pump intensity at the back of the crystal *)

\( \text{minimumsignal} = \text{Is}/.\text{sols[[1]]}[zL]; \) (* identifies the minimum signal intensity at the back of the crystal *)

\( \text{maximumpump} = \text{Ip}/.\text{sols[[1]]}[z0]; \) (* identifies the maximum pump intensity at the back of the crystal *)

\( \text{maximumsignal} = \text{Is}/.\text{sols[[1]]}[z0]; \) (* identifies the maximum signal intensity at the back of the crystal *)

\( \text{AppendTo}[\text{ΔODfromAlpha}, \text{ΔODList}]; \) (* Puts the values of \( \Delta \text{OD} \) into the empty list *)

\( \text{Dimensions}[\text{ΔODfromAlpha}] \) (* The first component in the list represents the \( \alpha \) values and the second component represents the \( \Delta \text{OD} \) *)

\( \text{ΔODfromAlpha} \)

\{1777.84\}

\{\{1.20389\}, \{1.20389\}\}

Plot all of the values on the same plot

\( \text{ListPlot}[\text{ΔODfromAlpha}, \text{PlotLegends} \to \text{SwatchLegend[\{freq\]}]]\) (* This appears to be plotting index values of \( \Delta \text{OD} \) for x-axis and actual values of \( \Delta \text{OD} \) for y-axis. I want to plot real values of \( \alpha \) or frequency for x-axis and real values of \( \Delta \text{OD} \) for y-axis. *)

\( \text{ΔODinRowMatrix} = \text{First}@\text{ΔODfromAlpha} \) (* Takes the *)

\( \text{FreqVSΔOD} = \text{Transpose}[\{\text{freq, ΔODinRowMatrix}\}]; \) (* Creates an array where the # of rows is determined by the # of \( \alpha \) values. The first column represents frequency or alpha, whichever you choose, and the rest of the columns are the \( \Delta \text{OD} \) values, which come from varying \( \alpha \) and E0. *)

\{1.20389,1.20389\}

\{785,1.20389\}, \{786,1.20389\}\}

\( \text{ListPlot}[\text{FreqVSΔOD, AxesLabel} \to \{\text{"Frequency", "ΔOD"}, \text{AxesStyle} \to \{\text{GrayLevel[0], Bold}\}]; \) (* This is frequency on the x-axis and actual \( \Delta \text{OD} \) values for y-axis *)
NewODvAlpha = Table[{freq[[i]], ΔODfromAlpha[[i]]}, {i, 1, Length[freq]}] (* Another way to connect frequency with ΔOD values like FreqVSΔOD above *)

(* Now I want to use Gary's code to model the actual grating formation of transmission (y-axis) vs time (x-axis). The code appears to be in Gary's Mathematica binder. I've labeled the useful parts with sticky notes *)

Transient Solution

(* \[Subscript[\epsilon, o]\] Permittivity Of Free Space; *)

\[\epsilon_o = \epsilon_0;\]

\[\epsilon_{rs} = \epsilon_r;\]

γ = 1; (* Electron-donor recombination coefficient *) (* CHECK THIS VALUE FOR KNbO3 *)

\[\tau_{diMax} = (\epsilon_r \epsilon_0) / ((q \mu s (minimumpump - minimumsignal) 1 *(ND - NA)/(\gamma NA) + (q \mu s Id*(ND - NA))/(\gamma NA)); (* Might have an issue with inability to determine if character is the number "1" or the letter "l" *) (* Estimate of the maximum value of the dielectric relaxation time (using beam intensities at the front of the crystal) *)

\[\tau_{diMin} = (\epsilon_r \epsilon_0) / ((q \mu s (maximumpump - maximumsignal) 1 *(ND - NA)/(\gamma NA) + (q \mu s Id*(ND - NA))/(\gamma NA)); (* Might have an issue with inability to determine if character is the number "1" or the letter "l" *) (* Estimate of the minimum value of the dielectric relaxation time (using beam intensities at the back of the crystal) *)

\[\Delta t = \tau_{diMin} / 1000; (* Time step for the transient solution in seconds. This method of dividing the estimated dielectric relaxation time by a denominator ensures that the time step unit will always be sufficiently small *)

\[\Gamma P = (2 \pi) / \lambda n^3 \text{reffP} \text{Im}((E0 + I*ED + EPV)/(1 + (ED - I*E0)/EQ - I (E0/EQ + NA/ND*EPV/EQ)); (* Gain coefficient for light as a function of the crystal angle with respect to the crystal c-axis *)

*aP =; (* a coefficient for the space charge field differential equation, for p-polarized light *)

*bP =; (* b coefficient for the space charge field differential equation for p-polarized light. *)

*cP =; (* c coefficient for the space charge field differential equation, for p-polarized light. *)

\[s = 1.623 \times 10^{-3}; (* photoexcitation cross section in m^2/W. Check this value! *)

n_α = 2.1815; (* α-axis refractive index for potassium niobate *)

n_β = 2.2958; (* β-axis refractive index for potassium niobate *)

n_γ = 2.3489; (* γ-axis refractive index for potassium niobate *)

nP = n_γ; (* ((Cos[\theta*Degree]^2/n_γ^2+(Sin[\theta*Degree]^2/n_β^2+(\gamma^{2}-\gamma^{2})^2)/(-1/2) Re refractive index with angle for p-polarized light. Did I choose the correct index??? *)

reffP =; (* I need to figure out how to find the effective pockels coefficient r for different polarizations *)

reffS =; (* I need to figure out how to find the effective pockels coefficient r for different polarizations *)
\( q^P = \pi/\lambda * n_P^3 * \text{reff}_P; \quad \) (* \( q \) coefficient for p-polarized light as a function of the crystal angle w.r.t the crystal c-axis *)

\[
\text{initialpump} = \text{FunctionInterpolation}[(1 - \text{Rf}) \times \text{pump} \times e^{-\alpha z}, \{z, 0, L\}] \quad (* \text{Initial pump intensity through the crystal at time } t=0 *)
\]

\[
\text{initialsignal} = \text{FunctionInterpolation}[((n-1)/(n+1))^2 \times (1 - \text{Rf}) \times \text{pump} \times e^{\alpha(z-2L)}, \{z, 0, L\}]; \quad (* \text{Initial signal intensity through the crystal at time } t=0 *)
\]

\[
\Delta \text{ODP} = \quad (* \text{Begins the transient solution calculation} *)
\]

\[
\text{multiplier} = 0; \quad (* \text{Sets the time step counter to zero} *)
\]

\[
\text{EscPrevious} = 0; \quad (* \text{Sets the space charge array to zero for time } t=0 *)
\]

\[
\text{transientlist} = \{0, 0\}; \quad (* \text{sets up the first elements of an array of time and } \Delta \text{OD values} *)
\]

\[
\text{steadystatelist} = \{\tau_{diMax}, \Delta \text{ODSteadyStateP}\}; \quad (* \text{Defines the start of the steady state list} *)
\]

\[
\text{pumpbeamdistribution} = \{0, \text{initialpump}\}; \quad (* \text{initial array of values for time and pump intensity distribution} *)
\]

\[
\text{signalbeamdistribution} = \{0, \text{initialsignal}\}; \quad (* \text{initial array of values for time and signal intensity distribution} *)
\]

\[
\text{steadystatepumpdistribution} = \{\tau_{diMax}, (Ip/.\text{solution}[1])\}; \quad (* \text{steady state pump beam intensity distribution} *)
\]

\[
\text{steadystatesignaldistribution} = \{\tau_{diMax}, (Is/.\text{solution}[1])\}; \quad (* \text{steady state signal beam intensity distribution} *)
\]

\[
\text{while} [\text{multiplier} \times \Delta t < \text{time}, (* \text{while loop repeats the calculation in time steps of } \Delta t \text{ until the value of } "\text{Time}" \text{ is reached} *)
\]

\[
\text{multiplier} = \text{multiplier}^1.001 + 1; \quad (* \text{Increments the time step by 1, gradually increasing the time step as the total value of time is increased carries a risk of accumulated numerical errors which become more pronounced for longer crystal lengths and higher gain values. However, without this time step expedient, the calculations would take an unreasonable amount of time. For example, with a total Fe concentration of 0.07 and an absorption coefficient of 100/cm, with a 5mm crystal, the minimum dielectric relaxation time is about 10msec. With a time step of 1/10 of the value (fairly course!) about 10,000 linear time iterations would be needed to reach steady state. Using an interative time exponent of 1.05 reduces the necessary time steps to about 190, making the calculations 50 times faster. The value of the temporal exponent can be reduced for better accuracy at the expense of calculations speed. Likewise, the denominator used to set } \Delta t \text{ can be increased for better accuracy with longer computation if required.} *)
\]

\[
\text{If}[\text{multiplier} \times \Delta t \geq \tau_{diMax}, \text{steadystatelist} = \text{Append}[\text{steadystatelist}, \{\Delta t \times \text{multiplier}, \Delta \text{ODSteadyStateP}\}]\], (* If statement uses the steady state solution if the value of "time" is greater than the largest possible value of the dielectric relaxation time, otherwise uses the full transient solution *)
\]

\[
\text{accuracy2} = \text{accuracy}; \quad (* \text{Calculation accuracy for the iterative loop} *)
\]

\[
\text{error2} = 1; \quad (* \text{Preliminary error value for the iterative loop calculation} *)
\]

\[
\text{Ipump} = \text{pump}; \quad (* \text{Ipump is the pump intensity at the crystal front face in W/m}^2 \text{ *)}
\]

\[
\text{MaximumReflection} = \text{Ipump} \times (1 - \text{Rf}); \quad (* \text{Maximum possible reflected intensity from the photorefractive grating in W/m}^2 \text{ *)}
\]

\[
\text{MinimumReflecfion} = 0; \quad (* \text{Minimum possible reflected pump intensity in W/m}^2 \text{ *)}
\]
count2=0; (* sets the iteration counter to 0 *)
while[Abs[error2]>accuracy2; (* While loop iterates the coupled intensity
equations down to a stable solution for each value of time *)
count2=count2+1; (* increments the counter *)
guess2=(1/2)*(MaximumReflection+MinimumReflection); (* Initial guess for
grating reflectivity *)
solution2=NDSolve[ (* DE solver for the coupled intensity equations *)
{Ip'[z]==-α*Ip[z]-Abs[2*qP*Ip[z]*Is[z]]/(Ip[z]*Is[z]*Id)*Im[EscPrevious]], (*
Differential equation for the pump beam *)
Is'[z]==α*Is[z]-Abs[2*qP*Ip[z]*Is[z]]/(Ip[z]*Is[z]*Id)*Im[EscPrevious]], (*
Differential equation for the signal beam *)
{Ip[0]==Ipump*(1-Rf),
Is[0]==guess2}, (*Initial boundary conditions for the differential equation
solutions *)
{Ip, Is}, (* Instruction to solve for the pump and signal beams and the phase *)
{z,0,L}, (* Integration range for the calculations *)
AccuracyGoal-> 12,
PrecisionGoal-> 12,
MaxStepSize-> L/10000,
MaxSteps-> Infinity];
desiredsignal=(Ip/.solution2[[1]])[L]*Rb; (* desired signal strength *)
actualsignal=(Is/.solution2[[1]])[L]; (* actual signal strength *)
If[count2>maxloops, error2=accuracy2, error2=(desiredsignal-
actualsignal)/actualsignal]; (* error definition within the iterative loop. The value
gradually diminishes until it meets the required solution accuracy. If the number
of iteration loops becomes higher than the pre-set maximum, then the calculation terminates
and uses the solution obtained after the pre-set maximum number of iteration loops *)
which[error2<0, MaximumReflection=guess2, error2>0, MinimumReflection=guess2];
(* End of inner While loop *)
pumpbeam2=(Ip/.solution2[[1]]);
signalbeam2=(Is/.solution2[[1]]);
escPrevious=EscPrevious* bP*Δt*multiplier
\[\frac{q*μ*σ*(ND-NA)}{γ*NA} \cdot (pumpbeam2[z]+signalbeam2[z]+Id)\] +cP/bP*(
\[\frac{q*μ*σ*(ND-NA)}{γ*NA} \cdot (pumpbeam2[z]+signalbeam2[z]+Id)\]
APPENDIX B

ESTABLISHED METHOD FOR CRYSTAL ANALYSIS

The following steps are for two-beam counter propagation measurements.

1. Turn on laser and wait for it to equilibrate. This may take up to an hour depending on the laser.

2. Choose a KNbO₃ or LiNbO₃ crystal which you would like to study.

3. A brown KNbO₃ crystal means that it has been reduced and is good to work with.

4. Place crystal on stage with “c+” axis in the direction of initial propagation.
   a. If there is a dot on the crystal it usually indicates the backside of the “c” axis.

5. For KNbO₃ cleavage planes are usually visible along the “b” axis.

6. The beam should be highly collimated before it enters the crystal stage lens, thus the focal position will be exactly at the back-focal length of the lens.

7. Place a power meter detector before the crystal stage to determine laser power (watts). Open shutter. Adjust polarizers to modulate laser to a very low power (i.e. ~20 µW). Close shutter and remove detector.

8. Open shutter and allow the low power laser beam to impinge on the crystal and look for how much light passes through due solely to reflection and absorption and record it. Close shutter.
9. Place detector in front of crystal stage again. Open the shutter and adjust the laser to the desired intensity. Close shutter and remove detector.

10. Turn on the oscilloscope and connect a power meter detector to it. Push the “trigger” button on the oscilloscope.

11. Use a 50\(\Omega\) terminator for quicker response times or 1K\(\Omega\) for more sensitivity

12. Place this detector behind the collimating lens which itself is behind the crystal stage. Open shutter and observe the characteristic \(\Delta OD\) pattern. Use the oscilloscope cursors to determine how high the voltage rises and record it. This is \(V_{\text{max}}\). Wait at least 1 minute and push the “trigger” button. You should see a mostly flat line. Use the cursors to find a reasonable average value for this line and record it. This is \(V_{\text{min}}\).

13. To find the change in optical density use, \(\Delta OD = \log\left(\frac{V_{\text{max}}}{V_{\text{min}}}\right)\)

14. A high \(\Delta OD\) means better deflection of the beam.

15. Repeat the process approximately 20 times and average the \(\Delta OD\)
APPENDIX C

MATLAB CODE

Code I – The codes for calculating ΔOD at the other wavelengths and powers are very similar to the code provided below and as such only one example is provided.

%% KNbO3 Measurements, VLOC K83B2-3, 2000 ppm - F=199.5 mm, F#80, Acquired 18July2017
%% Wavelength 532nm, Power 5mW
clear all; close all; clc;

%% Area 2 - Short time frame
KNbO3_Area1_zero=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\SHORT KNbO3 - K83B2-3 - Area2 - 532nm - 5mW.csv',0,3);
KNbO3_Area1=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\SHORT KNbO3 - K83B2-3 - Area2 - 532nm - 5mW.csv',312,3);
temp1=KNbO3_Area1_zero(:,2);
real_zero1=abs(mean(temp1(1:311)));%Average of data points when no light is entering the detector

fit_time1_K83B2=KNbO3_Area1(312:end,1);
fit_voltage1_K83B2=KNbO3_Area1(312:end,2);
figure(1)
plot(KNbO3_Area1(:,1),KNbO3_Area1(:,2))
title({'KNbO3, VLOC K83B2-3, Area 2-Short, Focus 199.5mm,' 'Incident 5mW, \lambda=532 nm'})
xlabel('Time (seconds)')
ylabel('Voltage (Volts)')

[MaxPeak1,maxIdx1]=max(KNbO3_Area1(:,2));
KNbO3_Area1(maxIdx1,1); %This is the time that the peak occurs
KNbO3_Area1_SS=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\SHORT KNbO3 - K83B2-3 - Area2 - 532nm - 5mW-SS.csv',0,3);
Low1_SS=mean(KNbO3_Area1_SS(:,2)); %Average of steady state data points
%[LowSpot1,lowIdx1]=min(KNbO3_Area1(:,2));
%Delta_OD_Area1=log10(MaxPeak1/LowSpot1)
Delta_OD_Area2_short_SS=log10((real_zero1+MaxPeak1)/(real_zero1+Low1_SS))

Ydecay1=KNbO3_Area1(:,2);
Xdecay1=KNbO3_Area1(:,1);
decay_power1=Low1_SS+(MaxPeak1-Low1_SS)*0.01; %This sets the curve fit end power
[poop1 index1]=min(abs(Ydecay1-decay_power1));
decay_power_closest_value1=Ydecay1(index1);
decay_time1=Xdecay1(index1)

%%  Area 2 - Long time frame
KNbO3_Area2_zero=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\LONG KNbO3 - K83B2-3 - Area2 - 532nm - 5mW.csv',0,3);
KNbO3_Area2=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\LONG KNbO3 - K83B2-3 - Area2 - 532nm - 5mW.csv',6220,3);
temp2=KNbO3_Area2_zero(:,2);
real_zero2=abs(mean(temp2(1:6219)));%Average of data points when no light is entering the detector

fit_time2=KNbO3_Area2(6221:end,1);
fit_voltage2=KNbO3_Area2(6221:end,2);
figure(2)
plot(KNbO3_Area2(:,1),KNbO3_Area2(:,2))
title({'KNbO3, VLOC K83B2-3, Area 2-Long, Focus 199.5mm,' 'Incident 5mW, \lambda=532 nm'})
xlabel('Time (seconds)')
ylabel('Voltage (Volts)')

[MaxPeak2,maxIdx2]=max(KNbO3_Area2(:,2));
KNbO3_Area2(maxIdx2,1); %This is the time that the peak occurs

KNbO3_Area2_SS=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\LONG KNbO3 - K83B2-3 - Area2 - 532nm - 5mW-SS.csv',0,3);
Low2_SS=mean(KNbO3_Area2_SS(:,2)); %Average of steady state data points
%[LowSpot2,lowIdx2]=min(KNbO3_Area2(:,2));
%Delta_OD_Area2=log10(MaxPeak2/LowSpot2)
Delta_OD_Area2_long_SS=log10((real_zero2+MaxPeak2)/(real_zero2+Low2_SS))

Ydecay2=KNbO3_Area2(:,2);
Xdecay2=KNbO3_Area2(:,1);
decay_power2=(Low2_SS+real_zero2)+((MaxPeak2+real_zero2)-(Low2_SS+real_zero2))*0.01; %This sets the curve fit end power
[poop2 index2]=min(abs(Ydecay2-decay_power2));
decay_power_closest_value2=Ydecay2(index2);
decay_time2=Xdecay2(index2)

%%  Area 3 - Short time frame

KNbO3_Area3_zero=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\SHORT KNbO3 - K83B2-3 - Area3 - 532nm - 5mW.csv',0,3);
KNbO3_Area3=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\SHORT KNbO3 - K83B2-3 - Area3 - 532nm - 5mW.csv',312,3);
temp3=KNbO3_Area3_zero(:,2);
real_zero3=abs(mean(temp3(1:310)));%Average of data points when no light is entering the detector

figure(3)
plot(KNbO3_Area3(:,1),KNbO3_Area3(:,2))
title({'KNbO3, VLOC K83B2-3, Area 3-Short, Focus 199.5mm,' 'Incident 5mW, \lambda=532 nm'})
xlabel('Time (seconds)')
ylabel('Voltage (Volts)')

[MaxPeak3,maxIdx3]=max(KNbO3_Area3(:,2));
KNbO3_Area3(maxIdx3,1); %This is the time that the peak occurs

KNbO3_Area3_SS=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\SHORT KNbO3 - K83B2-3 - Area3 - 532nm - 5mW-SS.csv',0,3);
Low3_SS=mean(KNbO3_Area3_SS(:,2)); %Average of steady state data points
%[LowSpot3,lowIdx3]=min(KNbO3_Area3(:,2));
%Delta_OD_Area3=log10(MaxPeak3/LowSpot3)
Delta_OD_Area3_short_SS=log10((real_zero3+MaxPeak3)/(real_zero3+Low3_SS))
%% Area 1 - Long time frame
KNbO3_Area4_zero=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\LONG KNbO3 - K83B2-3 - Area1 - 532nm - 5mW.csv',0,3);
KNbO3_Area4=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\LONG KNbO3 - K83B2-3 - Area1 - 532nm - 5mW.csv',6221,3);
temp4=KNbO3_Area4_zero(:,2);
real_zero4=abs(mean(temp4(1:6219)));%Average of data points when no light is entering the detector

figure(4)
plot(KNbO3_Area4(:,1),KNbO3_Area4(:,2))
title({'KNbO3, VLOC K83B2-3, Area 1-Long, Focus 199.5mm,' 'Incident 5mW, 
\lambda=532 nm'})
xlabel('Time (seconds)')
ylabel('Voltage (Volts)')

[MaxPeak4,maxIdx4]=max(KNbO3_Area4(:,2));
KNbO3_Area4(maxIdx4,1); %This is the time that the peak occurs

KNbO3_Area4_SS=csvread('C:\Users\micha\Graduate School\USAF\Research\KNbO3 Oscilloscope Data\18Jul2017\532nm, 5mW\LONG KNbO3 - K83B2-3 - Area1 - 532nm - 5mW-SS.csv',0,3);
Low4_SS=mean(KNbO3_Area4_SS(:,2)); %Average of steady state data points
%

Delta_OD_Area4=log10(MaxPeak4/LowSpot4)
Delta_OD_Area1_long_SS=log10((real_zero4+MaxPeak4)/(real_zero4+Low4_SS))

Ydecay3=KNbO3_Area3(:,2);
Xdecay3=KNbO3_Area3(:,1);
decay_power3=Low3_SS+(MaxPeak3-Low3_SS)*(1/exp(1)); %This sets the curve fit end power
[end power
[poop3 index3]=min(abs(Ydecay3-decay_power3));
decay_power_closest_value3=Ydecay3(index3);
decay_time3=Xdecay3(index3)

Code II

%% Calculating laser Powers
close all; clear all; clc;
nm532=5; %Units of mWatts
AC532=1.91312; %Units of 1/cm
AC447=3.22715; %Units of 1/cm
AC523=2.00354; %Units of 1/cm
AC561=1.60495; %Units of 1/cm
AC633=0.82136; %Units of 1/cm
AC785=0.16834; %Units of 1/cm

%Calculating the power adjustment needed to ensure we have approx. the same
%number of electrons and holes being excited across all wavelengths. This
%assumes a linear relationship between power and absorption coefficient.

scale447=AC532*nm532/AC447 %Units of mWatts
scale523=AC532*nm532/AC523 %Units of mWatts
scale561=AC532*nm532/AC561 %Units of mWatts
scale633=AC532*nm532/AC633 %Units of mWatts
scale785=AC532*nm532/AC785 %Units of mWatts

%%% Room temperature electron energy in crystal affecting hole creation
Kb=8.6173303e-5; %Units of eV/K
T=295; %Units of Kelvin
room_energy=Kb*T

%The band gap should shift by k to the right, but still need to
%find an accurate band gap diagram to know exactly how many eV it will
%lower by. This is important, because 447nm (2.77eV) is off from the
%2.35eV by 0.42eV and unless the room temperature energy can shift the
%crystal gap down by 0.42eV, which is unlikely, we won't be creating any holes.
lambda=1.24/room_energy
k=2*pi/lambda

**Code III**

%%% Focusing a Gaussian beam
clear all; close all; clc;

f200=0.200; %focal length in m
lambda532=532e-9; %units are m

Verdi_beam_diam=0.00234; %Beam diameter found using Cinogy camera. Units of m
W0=Verdi_beam_diam/2; %Beam waist radius. Units of m
Msquared_perfect=1;
Wnew\_measured=(70e-6)/2; %Measured Beam waist radius in m using Cinogy camera

Msquared\_measured=Wnew\_measured*sqrt(1+(f200*lambda532/(pi*W0^2))^2)/(f200*lambda532/(pi*W0))

Wnew\_rigorous\_ideal=(Msquared\_perfect*f200*lambda532/(pi*W0))/sqrt(1+(f200*lambda532/(pi*W0^2))^2) %Units are m. Assuming Msquared of 1
%Comparing rigorous formula to simplified form shows a high degree of agreement
Wnew\_simple\_ideal=Msquared\_perfect*f200*lambda532/(pi*W0) %Units are m. Assuming Msquared of 1
Wnew\_simple\_measured=Msquared\_measured*f200*lambda532/(pi*W0) %Units are m. Assuming Msquared of 1

RayleighRange\_ideal=pi*Wnew\_rigorous\_ideal^2/(lambda532*Msquared\_perfect)*1000 %Units are mm
RayleighRange\_measured=pi*Wnew\_measured^2/(lambda532*Msquared\_measured)*1000 %Units are mm
confocal\_ideal=2*RayleighRange\_ideal
confocal\_measured=2*RayleighRange\_measured

n\_crystal=2.3813;
n\_air=1.00029;
Reflection\_front=100*((n\_crystal-n\_air)/(n\_crystal+n\_air))^2; %Percent of reflection from surface
Reflection\_back=100*((n\_air-n\_crystal)/(n\_air+n\_crystal))^2; %Percent of reflection from surface

*Code IV*

%%% Retrapping Model and its Thermodynamic Implications

clear all; close all; clc;

Kb=8.6173303e-5; %Boltzmann Constant. Units of eV/K
Av=6.022e23; %Avogadro's Number. Units of atoms/mole
Ener=1.109; %Energy dissipated when "hot" electrons from 633nm, 1.959eV %move from high in the conduction band to the bottom of the CB. Units of eV
Temp=Ener/Kb %This is the temperature 1.109eV gives off
Percent\_electrons=0.15745/(0.50068+0.15745) %According to Origin's curve %fitting parms only ~25% of the electrons are being retrapped
Fe\_atoms=Av*1e-6 %Number of Fe atoms participating in retrapping
x=Temp*Fe\_atoms %Units of Kelvins/mole
y=x/Av %Units of Kelvin/atom
%To remove atom-1 just go back to thermo dynamics and take the ensemble
%average of the whole system and you are left with units of Kelvin

%If we have a best case scenario of only one retrapping occurring, then
%the lifetime tau2 is the amount of time to bleed off this 1.109eV
tau2=0.01025;
z=y/tau2 %Units of Kelvin/sec

%If retrapping is really occurring then that means at 633nm 1.1eV has to
%bleed off with every retrapment/jump. The crystal would heat up way too
%much if this were true. And there would be more luminescence as the
%electrons decay 0.85eV down into the Fermi level.

\textit{Code V}

%% Testing Curve Fit - 447 nm, 3mW, 6Jun2017
clear all; close all; clc;

%% Three Exponentials
%% Using real values for first two coefficients and exponentials. The third
%% is made up.
A0=0.0006; A1=0.01329; A2=0.00211
A3vers1=-0.001; A3vers2=-0.0005; A3vers3=-0.0001;
tau1=0.0028; tau2=0.03096
tau3vers1=0.3096; tau3vers2=1.0; tau3vers3=3.096;
t=0:0.00001:1;

a1=A0+A1.*exp(-t./tau1)+A2.*exp(-t./tau2)+A3vers1.*exp(-t./tau3vers1);
a2=A0+A1.*exp(-t./tau1)+A2.*exp(-t./tau2)+A3vers2.*exp(-t./tau3vers1);
a3=A0+A1.*exp(-t./tau1)+A2.*exp(-t./tau2)+A3vers3.*exp(-t./tau3vers1);

figure(1);
plot(t,a1,'r')
hold on;
plot(t,a2,'k')
hold on;
plot(t,a3,'g')
xlim([-0.1 1])
legend('A3=-0.00143','A3=-0.0008', 'A3=-0.000143')

b1=A0+A1.*exp(-t./tau1)+A2.*exp(-t./tau2)+A3vers1.*exp(-t./tau3vers1);
b2=A0+A1.*exp(-t./tau1)+A2.*exp(-t./tau2)+A3vers1.*exp(-t./tau3vers2);
b3 = A0 + A1 \cdot \exp(-t./\tau1) + A2 \cdot \exp(-t./\tau2) + A3 \cdot \exp(-t./\tau3) \\

figure(2); plot(t,b1,'r'); hold on; plot(t,b2,'k'); hold on; plot(t,b3,'g'); xlim([-0.1 1]); legend('\tau3=0.05264', '\tau3=0.3', '\tau3=5.264')
APPENDIX D

THERMODYNAMIC ANALYSIS OF RETRAPPING MODEL

For simplicity look only at 633 nm curve fitting values.

<table>
<thead>
<tr>
<th>Origin Curve Fitting Parameters</th>
<th>Power (mW)</th>
<th>Wavelength</th>
<th>Lifetimes (sec.)</th>
<th>Coefficient</th>
<th>ΔOD</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>11.96</td>
<td>633</td>
<td>0.00112 τ₁</td>
<td>0.50068 A₁</td>
<td>1.3816</td>
</tr>
<tr>
<td></td>
<td>1.959 eV</td>
<td>0.01025 τ₂</td>
<td>0.15745 A₂</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Starting with 2000ppm in the melt for this particular iron-doped crystal, we expect to have only around 20 ppm in the fully formed crystal. According to Origin’s curve fitting parameters about 25% of excited electrons from Fe²⁺ are retrapped, (0.15745/(0.50068+0.15745)=0.239). Assuming a fairly low number of the 20 ppm iron atoms are Fe²⁺ and of those only about 25% participate. With these assumptions, one can estimate that 1 ppm are participating in retrapping. In other words, multiply Avogadro’s number by 1e-6 and you obtain 6.022e17 atoms participating in retrapping. Using the energy schematic in Figure 1.1 means that if 633 nm (1.959 eV) is impinging on crystal then the electrons are 1.1 eV into the conduction band. Assume a best case scenario of only one retrapping occurring and that means 1.1 eV has to bleed off with every retrapped electron. *(Side note: IR luminescence should increase somewhat, because they would then give off 0.85 eV to fall back to the Fe³⁺ level.)* Which is equivalent to 12,869
Kelvin. Multiply the temperature by the number of participating atoms and you obtain 7.75e21 K/mol. Divide that value by Avogadro’s number to arrive at 0.0129 K/atom. To remove the units of atom⁻¹ just go back to thermodynamics and take the ensemble average of the whole system and you are left with units of Kelvin. As mentioned earlier assume a best case scenario of only one retrapping occurring, then the lifetime τ₂ is the amount of time the electron has to bleed off 1.109eV (0.0129K). So divide 0.0129K/0.01025=1.3K/sec. The laser is focused to approximately 70 microns and heating it by 1.3K/sec in that small of an area would almost certainly crack the crystal.