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THE INTERFEROMETRIC MEASUREMENT OF PHASE MISMATCH IN
POTENTIAL SECOND HARMONIC GENERATORS

The University of Arizona

Ph.D. 1984

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THE INTERFEROMETRIC MEASUREMENT OF PHASE MISMATCH
IN POTENTIAL SECOND HARMONIC GENERATORS

by

Edward Lawrence Sinofsky

A Dissertation Submitted to the Faculty of the
COMMITTEE ON OPTICAL SCIENCES (GRADUATE)

In Partial Fulfillment of the Requirements
For the Degree of

DOCTOR OF PHILOSOPHY

In the Graduate College

THE UNIVERSITY OF ARIZONA

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As members of the Final Examination Committee, we certify that we have read
the dissertation prepared by Edward Lawrence Sinofsky
entitled The Interferometric Measurement of Phase Mismatch in Potential
Second Harmonic Generators

and recommend that it be accepted as fulfilling the dissertation requirement
for the Degree of Doctor of Philosophy

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SIGNED: Edward Laurence Snodgrass

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ABSTRACT

This dissertation combines aspects of lasers, nonlinear optics and interferometry to measure the linear optical properties involved in phase matched second harmonic generation,(SHG). A new measuring technique has been developed to rapidly analyze the phase matching performance of potential SHGs. The data taken is in the form of interferograms produced by the self referencing nonlinear Fizeau interferometer (NLF), and correctly predicts when phase matched SHG will occur in the sample wedge.

Data extracted from the interferograms produced by the NLF, allows us to predict both phase matching temperatures for noncritically phase matchable crystals and crystal orientation for angle tuned crystals. Phase matching measurements can be made for both Type I and Type II configurations.

Phase mismatch measurements were made at the fundamental wavelength of 1.32 μm . for: calcite, lithium niobate, and gadolinium molybdate (GMO). Similar measurements were made at 1.06 μm . for calcite. Phase matched SHG was demonstrated in calcite, lithium niobate and KTP, while phase matching by temperature tuning is ruled out for GMO.

CHAPTER 1

INTRODUCTION

Soon after the introduction of the ruby laser, Peter Franken et al.(1961), suggested that if they irradiated certain crystals with the intense red light available from this laser, blue light could be generated by a process now known as second harmonic generation. Franken's group proceeded to demonstrate this process, marking the birth of nonlinear optics. The amount of blue light generated was very small because the interaction was not phase matched.

Maker et al.(1962), later showed that if the dephasing of the fundamental and second harmonic could be made to approach zero, the second harmonic field could grow quite large. This is commonly referred to as phase matching. Phase matching in crystals manifests itself through the linear property of refractive index. Thus, measurement of phase mismatch is a measurement of the material's linear optical properties, which are as important as the material's nonlinear optical properties for efficient SHG.

Maker achieved phase matching by using an anisotropic crystal, where the birefringence was adjusted to compensate for the dispersion, allowing the linear and nonlinear waves to propagate in phase with each other. Phase matched SHG was observed by orienting the crystal properly with respect to the incoming laser field. This is known as angle tuning. Initial experiments were performed by rotating a

crystal about two axes in the presence of an intense laser beam, while monitoring the second harmonic output.

In 1965 a paper by Miller et al., showed that phase matching could be achieved not only by angle tuning, but by carefully adjusting the crystal temperature to vary the crystal's dispersion curves. This is now known as noncritical phase matching (NCPM) because the angular accuracy needed for critical phase matching (CPM) is orders of magnitude more severe. Prior to the present work, searching for NCPM materials has involved the measurement of SHG as the crystal temperature is scanned. These experiments can be frustrating if the phase matching temperature cannot be reached by one's experimental apparatus. Information on whether to cool or heat the crystal, and by how much, is not obtained in experiments of this type.

The research described in this dissertation allows us to quantitatively measure the phase mismatch as crystal angle or temperature is varied, and avoids the need to make tedious measurements of refractive indices to within the fourth decimal place necessary to make accurate phase match calculations. By evaluating interferograms of the samples produced in the NLF, one can quickly tell whether to heat or cool the crystal, as well as permitting us to predict the phase matching temperature without ever reaching that temperature in a search for NCPM materials. In the case of angle tuned crystals, we obtain information on how to orient the crystal.

When using the technique of orthogonal dispersion, described later in the text, we can evaluate phase mismatch qualitatively by

observing fringe tilt. Orthogonal dispersion also calibrates the phase mismatch of the sample against a wedge of known dispersion, allowing quantitative analysis.

Alternative to the use in the generation of high power, short wavelength laser fields, some crystals with lower conversion efficiencies have been shown to be versatile diagnostic tools as components of nonlinear optical interferometers as described in Chapter 3, (Hopf and Cervantes 1982). One version of this interferometer, the nonlinear Fizeau, is used with collimated laser beams to search for crystals to be used in interferometers at $1.32\text{ }\mu\text{m}$. that can contour objects with arbitrarily curved surfaces. Although GMO is tested for a Type II NCPM for use as a spatial correlator, the unsuitability of this material means that the search has just begun. Measurements of angle tuned crystals are made for completeness, because some important new nonlinear materials might not temperature tune.

The interferometric measurement of phase mismatch is an important tool for use in future nonlinear optical materials searches. The ability to quickly and easily analyze materials for phase match will allow many new materials to be tested. With current research efforts aimed towards developing new organic materials for SHGs, there will be a need for the measuring technique described.

Chapter 2 is a review of concepts involved in the second harmonic generation process. These concepts are the key to the understanding of the described testing procedure. Chapter 3

introduces the concept of nonlinear interferometry, and explains how the NLF will be used in crystal testing, both in Type I and Type II configurations. Chapter 4 is a survey of the nonlinear optical materials that were tested. Test results, as well as comparisons between results and previously released data available are also presented here.

CHAPTER 2

GENERAL PRINCIPLES

Second harmonic generation is one of the many nonlinear optical phenomena that can be described accurately with the theory of classical electrodynamics. In this theory, matter is composed of nuclei and their surrounding charge clouds. Interaction with an electromagnetic field causes the electron clouds and nuclei to move in opposite directions. Since the nuclei are many orders of magnitude more massive than the charge clouds, the nuclei can be considered fixed, with the bound electron clouds free to oscillate. The sum of these induced charge separations (dipole moments) is known as the polarization. This polarization is usually linear in the applied field, and is written:

$$P_L = \epsilon_0 \chi E \quad (1)$$

If the electric field is a travelling wave, the induced charge oscillation creates a polarization wave inside the medium. The total field in the material is the vector sum of the incident and reradiated field due to the polarization wave. These two fields may have different phases. The phase lag between incident and reradiated fields cause interference "downstream" in the material. This interference manifests itself as the linear optical properties of refraction and absorption.

Certain classes of crystals, however, also exhibit a nonlinear response, in which polarization waves at some frequencies, can induce polarization waves at other frequencies. The nonlinear polarization is written:

$$P_{NL}(r,t) = \epsilon_0 \chi^{(2)} : E(r,t) E(r,t) \quad (2)$$

In the most general case, this quadratic nonlinearity allows three wave mixing in which two fields combine to produce a third. SHG is a degenerate case of three wave mixing, in which the two fields to be combined are from the same laser, with the third field at twice the laser frequency. The incident laser field is written:

$$E = \frac{1}{2} (E(\omega, k) e^{i(k \cdot z - \omega t)} + cc) \quad (3)$$

This field squares to give nonlinear polarizations that have DC terms, as well as a polarization wave at twice the fundamental frequency, caused by adding two fields at the fundamental frequency in a nonlinear medium.

Since the nonlinear polarization is a weak effect it can only be observed in the presence of intense laser light. Because of this, however, we can use perturbative approximations. The total polarization wave in the medium is written:

$$P = P_L + P_{NL} \quad (4)$$

where P_L is the linear polarization, and P_{NL} is the nonlinear response. This polarization acts as a source that drives the displacement field D via Maxwell's equation:

$$-\nabla^2 D + \frac{1}{c^2} \frac{\partial^2 D}{\partial t^2} = \nabla \times \nabla \times (P_L + P_{NL}) \quad (5)$$

If we assume P_L is a plane wave eigenvector, with a well defined index, n :

$$-\nabla^2 D + \frac{n^2}{c^2} \frac{\partial^2 D}{\partial t^2} = \nabla \times \nabla \times P_{NL} \quad (6)$$

These are vector equations that allow for two orthogonal eigenvectors with potentially different indices. We now specify the harmonic dependance of the displacement field, D , and allow the amplitude to vary slowly with propagation distance, z , by writing:

$$D = \frac{1}{2} (D(z; 2\omega, k) e^{i(kz - 2\omega t)} + cc) \quad (7)$$

where cc denotes complex conjugate. The weak polarization is written:

$$P_{NL} = \frac{1}{2} (P_{NL}(z; 2\omega, k_p) e^{i(k_p z - 2\omega t)} + cc) \quad (8)$$

We now invoke the slowly varying amplitude and phase condition (SVEA), which states that the field amplitudes must vary slowly over distances comparable to an optical wavelength. Mathematically, the condition reads:

$$\left| \frac{\partial D(z; 2\omega, k)}{\partial z} \right| \ll k |D(z; 2\omega, k)| \quad (9)$$

The curl-curl operation is now reexpressed with the projection operator, $\hat{O}(k_p)$ (Hopf and Stegeman, to be published), ignoring all spatial derivatives of P_{NL} .

$$\nabla \times \nabla \times P_{NL} = \frac{1}{2} (k_p^2 \hat{O}(k_p) \cdot P_{NL} e^{i(k_p z - 2\omega t)}) \quad (10)$$

We now evaluate the left side of (6) using (7):

$$-\nabla^2 D + \frac{n^2}{c^2} \frac{\partial^2 D}{\partial t^2} = \frac{1}{2} \left(-\frac{\partial^2 D}{\partial z^2} - 2ik \frac{\partial D}{\partial z} + (k^2 - \frac{n^2 \omega^2}{c^2}) D \right) e^{i(kz - 2\omega t)} + cc \quad (11)$$

Terms involving k^2 and $\frac{n^2 \omega^2}{c^2}$ are large, and must cancel. If we assume that the fundamental eigenvectors are only slightly perturbed by the nonlinearity, we can write:

$$k = \frac{n\omega}{c} \quad (12)$$

The second spatial derivative is small compared to $\left| 2ik \frac{\partial D}{\partial z} \right|$. Hence in the SVEA approximation:

$$-\nabla^2 D + \frac{n^2}{c^2} D = \frac{1}{2} (-2ik \frac{\partial D}{\partial z} e^{i(kz-2\omega t)} + cc) \quad (13)$$

Combining equations (6) and (10) one obtains:

$$-2ik \frac{\partial D}{\partial z} = k_p^2 O(k_p) \cdot P_{NL} e^{i(k_p - k) \cdot z} \quad (14)$$

where the second harmonic time dependence is temporarily suppressed. The quantity $k_p - k$ is known as phase mismatch. Measurement of this quantity is the fundamental tool used to perform the research for this dissertation. Phase mismatch is commonly written as:

$$\Delta k = k_p - k = (k_p - k) \hat{z} = \Delta k \hat{z} \quad (15)$$

Equation (14) can now be rewritten, splitting the vector equations into two scalar equations involving two orthogonal eigenvectors:

$$\frac{\partial D_a}{\partial z} = \frac{in_a \omega}{2c} \hat{e}_a \cdot \hat{O}(k) \cdot P_{NL} e^{i(\Delta k z)} \quad (16)$$

$$\frac{\partial D_b}{\partial z} = \frac{in_b \omega}{2c} \hat{e}_b \cdot \hat{O}(k) \cdot P_{NL} e^{i(\Delta k z)} \quad (17)$$

These equations are now expressed in terms of electric fields using:

$$E = \frac{D}{n^2 \epsilon_0} \quad (18)$$

Then:

$$\frac{\partial E_a}{\partial z} = \frac{i\omega}{2n_a \epsilon_0 c} \hat{e}_a \cdot \hat{O}(k) \cdot P_{NL} e^{i(\Delta k z)} \quad (19)$$

$$\frac{\partial E_b}{\partial z} = \frac{i\omega}{2n_b \epsilon_0 c} \hat{e}_b \cdot \hat{O}(k) \cdot P_{NL} e^{i(\Delta k z)} \quad (20)$$

The nonlinearity polarization of interest here is proportional to the square of the field.

$$P_{NL}(\mathbf{r}, t) = \epsilon_0 \chi^{(2)} : \mathbf{E}(\mathbf{r}, t) \mathbf{E}(\mathbf{r}, t) \quad (21)$$

Standard notation for this nonlinearity defines the d tensor in terms of the nonlinear susceptibility.

$$d \equiv \frac{1}{2} \chi^{(2)} \quad (22)$$

The factor of 2 is inserted so that in Levi-Civita notation:

$$P_{iNL}(2\omega) = \epsilon_0 d_{ijk} E_j(\omega) E_k(\omega) \quad (23)$$

Further reference on the tensorial nature of the nonlinear polarization can be found in Applied Nonlinear Optics by Midwinter and Zernicke.

The generation of the nonlinear wave involves projections of all the eigenvectors onto the d tensor. If the eigenvectors are associated with the three unit vectors; $\hat{e}_a$, $\hat{e}_b$, $\hat{e}_c$, we define:

$$d_{eff} \equiv (\hat{e}_c \cdot d) : \hat{e}_a \hat{e}_b \quad (24)$$

This is a contraction product between d and the three directions involved in the nonlinear interaction. Thus d_{eff} can be expressed as:

$$d_{eff} = \hat{e}_a \cdot \hat{O}(k) \cdot P_{NL} \quad (25)$$

In SHG there are two possible configurations. In a Type I configuration a second harmonic polarization wave is created from two fundamental eigenvectors polarized in the same direction. In a Type II configuration the second harmonic polarization wave is created from two orthogonally polarized eigenvectors. One can see this more clearly by writing the total field just inside the crystal, at $z = 0$. For a Type I configuration:

$$E_f(r, \omega_f) = \frac{1}{2} (\hat{e}_f E(k_f, \omega_f) e^{i(k_f \cdot r)} + \text{c.c.}) \quad (26)$$

For a Type II configuration:

$$E_f(r, \omega_f) = \frac{1}{2} (\hat{e}_o E_o(k_{of}, \omega_f) e^{i(k_{of} \cdot r)} + \hat{e}_e E_e(k_{ef}, \omega_f) e^{i(k_{ef} \cdot r)} + \text{c.c.}) \quad (27)$$

In the Type II case, the input consists of a an o-ray and and e-ray, polarized in the principle planes of the crystal.

For Type I SHG:

$$k_p = 2k_f \quad (28)$$

with the effective polarization amplitude written as:

$$P_{NL}(2\omega_f, k_p) = \epsilon_0 d_{eff} E_f^2 \quad (29)$$

In a Type II interaction:

$$k_p = k_{of} + k_{ef} \quad (30)$$

with:

$$P_{NL}(2\omega_f, k_p) = 2\epsilon_0 d_{eff} E_{of} E_{ef} \quad (31)$$

The factor of two accounts for degeneracy in the Type I interaction.

Equations 19 and 20 are now modified so that they describe the growth of the second harmonic field, E_s . For Type I interactions:

$$\frac{\partial E_s}{\partial z} = -\frac{i\omega}{n_s c \epsilon_0} d_{\text{eff}} E_f^2 e^{i(\Delta k z)} \quad (32)$$

For Type II interactions:

$$\frac{\partial E_s}{\partial z} = -\frac{i\omega}{2n_s c \epsilon_0} d_{\text{eff}} E_f^2 e^{i(\Delta k z)} \quad (33)$$

integrating from 0 to z we obtain:

$$E_s(z) = \frac{i\omega}{4n_s c \epsilon_0} d_{\text{eff}} E_f^2 z e^{i(\frac{\Delta k z}{2})} \frac{\sin \frac{\Delta k z}{2}}{\frac{\Delta k z}{2}} \quad (34)$$

from this expression the time averaged Poynting vector is calculated:

$$S_s = \frac{\omega^2}{32n_s c \epsilon_0} d_{\text{eff}}^2 E_f^4 z^2 \frac{\sin^2(\frac{\Delta k z}{2})}{(\frac{\Delta k z}{2})^2} \hat{z} \quad (35)$$

Except for $\Delta k = 0$, (phase matching), the second harmonic flux generated varies sinusoidally with distance, z. The distance where the SHG power reaches a maximum is called the coherence length, and is defined by:

$$L_{\text{coh}} = \frac{\pi}{\Delta k} \quad (36)$$

For an arbitrary Δk , L_{coh} is usually on the order of a few wavelengths of light, and the flux is extremely small. If the interaction is phase matched, however, the radiation can become quite large. In that case:

$$S_s = \frac{\omega^2}{32n_s c \epsilon_0} d_{\text{eff}}^2 E_f^4 z^2 \hat{z} \quad (37)$$

In general, Δk is not equal to 0. This is called phase mismatch. The associated sinusoidal variation in flux is due to interference effects. The light emitted at z is exactly out of phase

with the light emitted at $z + L_{\text{coh}}$. The research done for this dissertation interferometrically measures this phase mismatch, in a search for new phase matchable nonlinear optical materials.

In an optical material, the amplitude of the real part of the wave vector is given by:

$$k = \frac{n\omega}{c} = \frac{2\pi n}{\lambda} \quad (38)$$

For phase matching a Type I interaction:

$$2k_f = k_s \quad (39)$$

$$\frac{4\pi n_f}{\lambda_f} = \frac{2\pi n_s}{\lambda_s} \quad (40)$$

$$\lambda_s = \frac{\lambda_f}{2} \quad (41)$$

$$n_f = n_s \quad (42)$$

Phase matching for the Type I case is achieved when the fundamental and second harmonic fields see the same index, and propagate through the medium in phase.

In Type II phase matching:

$$k_{of} + k_{ef} = k_s \quad (43)$$

then:

$$\frac{n_{of} + n_{ef}}{2} = n_s \quad (44)$$

Phase matching in the Type II case occurs when the second harmonic field sees an index that is the average of the fundamental indices.

Crystal Optics

The classical electrodynamic model of the interaction of light with matter states that the charge cloud surrounding the nucleus is bound by a springlike force. The behavior of the charge cloud in the presence of an external light field is that of a damped harmonic oscillator. Crystal symmetries allow for the forces to be diagonalized in some coordinate system. This coordinate system is called the "principle axis system". Each axis can have a different "spring constant", allowing for plane polarized eigenvectors to be excited along these axes. Each spring constant can be related to the phase velocity, or index of refraction, of the polarized eigenvector.

Isotropic materials have the same spring constant along all three directions in the crystal. A typical dispersion curve for an isotropic material has only one branch. Isotropic materials cannot be phase matched because there is no way to get the fundamental and second harmonic eigenvectors to see the same index.

Certain classes of crystals, however, have different spring constants, hence different refractive indices, depending on the axis of polarization. Uniaxial crystals have two independent dispersion curves, resulting from the two different "spring" constants. Biaxial crystals have three branches of the dispersion curves, caused by three independent "spring" constants. Uniaxial and biaxial crystals can

often be phase matched if the birefringence, or difference in index between branches of the dispersion curves, exceeds the dispersion between fundamental and second harmonic indices.

Two types of eigenvectors can be excited in nonisotropic crystals. An "o" configuration eigenvector is created when the electric field is parallel to a principle axis. The phase velocity of the field is determined by the index associated with the direction of the E field, and the poynting vector S is coincident with the wave vector k. Both the electric field E and the displacement field D are parallel in an o configuration. An "e" configuration eigenvector is created when both E and k lie in a plane defined by two principle axes. The phase velocity, or index, the field experiences is determined by the field direction, and the poynting vector S is not in general parallel to k. Also, E is not in general parallel to D. Figure 1 illustrates the e configuration. Using the coordinate system shown in Figure 1 we can write:

$$\mathbf{k} = k(\sin\theta, 0, \cos\theta) \quad (45)$$

Since $\mathbf{D} \cdot \mathbf{k} = 0$, D has the form:

$$\mathbf{D} = D(\cos\theta, 0, -\sin\theta) \quad (46)$$

Relating D and E:

$$\mathbf{E} = D\left(\frac{\cos\theta}{\epsilon_1}, 0, -\frac{\sin\theta}{\epsilon_3}\right) \quad (47)$$

By using these relations and manipulating the wave equations, we can now solve for index seen by the E field as a function of n_1 , n_3 , and angle θ with respect to the three axis.

$$n(\theta) = \frac{n_1 n_3}{(n_3^2 \cos^2 \theta + n_1^2 \sin^2 \theta)^{1/2}} \quad (48)$$

Let us now calculate the angle δ between the E and D vectors which is also the angle between the S and k vectors. One proceeds as follows; compute $D \cdot E$ to get $\sin \delta$ and $D \cdot E$ to get $\cos \delta$. The ratio of these simplify to obtain:

$$\tan \delta = \frac{n^2(\theta)}{2} \left(\frac{1}{n_3^2} - \frac{1}{n_1^2} \right) \sin 2\theta \quad (49)$$

This angle is called the walkoff angle in nonlinear optics, and it causes detrimental beam separation in nonlinear optical processes.

Phase matching is accomplished by either manipulating the angle θ between principle axes, or carefully adjusting the crystal temperature to tune the crystal indices. The former is known as angle phase matching, while the latter is known as noncritical phase matching, or 90° phase matching.

Phase Matching in Anisotropic Crystals

Phase matching requirements can be satisfied when any two wave vectors add to produce the third wavevector. When the three wavevectors are nonparallel, the phase matching is noncollinear. The major problem with noncollinear phase matching is that the interaction length between the two beams is limited by the beam diameter. In collinear phase matching all the wavevectors are parallel. As shown previously, phase matching in nonisotropic media is really index matching, with Type I and Type II index matching conditions stated in eqns. 42 and 44.

In materials where the birefringence exceeds the dispersion, a Type I angle tuned phase match is typically obtained by making the fundamental eigenvector an o ray, with the second harmonic eigenvector an e ray. The second harmonic wavevector is angle tuned in the 1-3 principle plane until phase matching is achieved when:

$$n_{of} = n_{es}(\theta) \quad (50)$$

This type of phase matching is known as an ooe configuration because two o ray fundamentals mix to produce an e ray second harmonic.

A Type II interaction allows two different configurations. In an ooe configuration, one fundamental eigenvector is polarized along an e direction, while the other fundamental, along with the second harmonic, are polarized along the o direction. The ooe Type II index matching condition is given by:

$$\frac{n_{of} + n_{ef}(\theta)}{2} = n_{os} \quad (51)$$

In an ooe Type II configuration, one fundamental is an o ray, one is an e ray, and the second harmonic polarization is an e ray. The ooe Type II configuration is given by:

$$\frac{n_{of} + n_{ef}(\theta)}{2} = n_{es}(\theta) \quad (52)$$

The type of phase matching described above is also commonly referred to as critical phase matching (CPM). This is because of the extreme precision with which the phase match angle must be held. The critical angle is defined as the angle of tilt away from the phase match angle at which the crystal length is equal to the $\frac{l_{coh}}{2}$. We

first expand $n_{es}(\theta)$ about the phase match angle θ_{pm} . Then:

$$n_{es}(\theta) \approx n_{es}(\theta_{pm}) + \left. \frac{\partial n_{es}(\theta)}{\partial \theta} \right|_{\theta=\theta_{pm}} \Delta\theta \quad (53)$$

By taking the derivative of eqn. 48, simplifying, and solving for $\Delta\theta$, the critical angle, one obtains:

$$\Delta\theta \approx \frac{\lambda_f}{4l} [(n_{os} - n_{es}) \sin 2\theta_{pm}]^{-1} \quad (54)$$

Where λ_f is the fundamental vacuum wavelength, and l is the crystal length. Since curved wavefronts have different k vectors depending on location on the wavefront, one cannot efficiently double the whole wavefront. When focusing into a CPM SHG to get high efficiency due to the high electric fields, one notices "Maker fringes" which are intensity variations in the critical dimension (perpendicular to the angle tuned plane). A typical Maker fringe pattern is shown in Figure 2. Maker fringes can be useful to measure the radius of curvature of the fundamental laser. The radius of curvature of the input laser is given by :

$$R = \frac{\Delta x}{\Delta\theta} \quad (55)$$

Where Δx is the maker fringe spacing and $\Delta\theta$ is the critical angle. It should be noted that the highest sensitivity is obtained using CPM SHGs with large index differences, hence small critical angles. Although useful for collimating laser beams, Maker fringes are detrimental in nonlinear interferometry, where we want to double

Figure 2. Maker fringes.

entire wavefronts which have information encoded on them in the form of irregular or curved wavefronts.

In any if the above mentioned angle tuned configurations we must not forget the walkoff effect that affects any e ray. In a Type I ooe configuration the second harmonic eigenvector separates from the fundamental. In a Type II ooe configuration one of the fundamentals separates from the other, limiting the interaction length. In a Type III ooe configuration both the second harmonic along with the fundamental both separate from each other as well as the fundamental polarized along the o direction.

Both walkoff and maker fringes can be eliminated in the special case of $\theta_{pm} = 90^\circ$. This type of phase matching is called noncritical phase matching, 90° phase matching or temperature tuned phase matching. As the name suggests, index manipulation is accomplished by changing the crystal's temperature. Temperature is one of the most powerful way to change some material's indices. Changes in temperature cause some crystals to change their lattice spacing. Small changes in lattice spacing can cause a large change in the internal binding forces in the crystal which in turn can effect the eigenvector's index. For a Type I ooe NCPM the index condition reads:

$$n_{of}(T_c) = n_{es}(90^\circ, T_c) \quad (56)$$

Where T_c is the phase match temperature. From now on the 90° notation will be suppressed. It is extremely unlikely that this condition will be satisfied at room temperature. The index condition

is then rewritten to allow for temperature tuning.

$$n_{of}(T_0) + \frac{\partial n_{of}}{\partial T} \Delta T = n_{es}(T_0) + \frac{\partial n_{es}}{\partial T} \Delta T \quad (57)$$

Where T_0 is room temperature and $\Delta T = T_c - T_0$. The temperature tolerance of an NCPM is calculated just as in the case of the CPM.

The condition $l = l_{coh}$ implies:

$$\Delta T_c = \left(\frac{\lambda_f}{4l}\right)^{1/2} \left[\frac{\partial n_{es}}{\partial T} - \frac{\partial n_{of}}{\partial T} \Big|_{T = T_c} \right]^{-1} \quad (58)$$

Notice that if the crystal indices change readily with temperature the phase matching temperature tolerance is very tight and visa versa. The critical angle in NCPM crystals appear to approach infinity for $\theta = 90^\circ$. This is because we only kept first order terms in the Taylor expansion. The second order terms from the expansion of eqn 53 gives:

$$\Delta \theta_c = \left(\frac{\lambda_f}{2l}\right)^{1/2} \left(\frac{\partial^2 n}{\partial \theta^2} \Big|_{\theta = 90^\circ} \right)^{-1/2} \quad (59)$$

For a Type I configuration in a uniaxial crystal the critical angle works out to be:

$$\Delta \theta_c = \left[\frac{\lambda_f}{4l(n_{os} - n_{es})} \right]^{1/2} \quad (60)$$

This critical angle is about two orders of magnitude larger for an NCPM SHG than that for a CPM SHG. This is enough of an increase to entirely double reasonably curved wavefronts without producing Maker fringes. In chapter 3 we will present interferometers that use this feature to be able to contour nonplanar surfaces.

CHAPTER 3

NONLINEAR INTERFEROMETRY

During the past few years, researchers at the Optical Sciences Center, under the direction of Dr. Fred Hopf, have developed several different nonlinear interferometers. These interferometers have proved to be very versatile diagnostic tools whose full potential has not yet been exploited. These interferometers are sensitive to wavefront dispersion between the fundamental and second harmonic fields, producing an interference patterns in the second harmonic wavelength, (in our case the visible). No beamsplitters are needed, laser coherence is not a concern, fringes are separated as if a much longer wavelength was used, and they can capture events that occur in nanoseconds (billionths of a second). Each of these features make these interferometers extremely easy to use, allowing many of the stringent alignment, component quality and vibration isolation requirements of typical visible wavelength interferometers to be relaxed. Nonlinear optical interferometers have measured machined pieces, lenses, chemical reactions and optical fiber preforms. In this dissertation we examine the use of one of these interferometers, the nonlinear Fizeau (NLF) to analyze possible phase matchable SHGs in a materials search. The NLF as well as some of the other nonlinear interferometers are described in detail in the dissertations of Cervantes (1982) and Liepmann (1983).

The NLF interferometer is setup by sandwiching some dispersive object or event between two phase matched SHGs (Figure 3,4).

The first SHG produces a small amount of second harmonic which then propagates collinearly with the higher power fundamental. These two wavefronts are then passed through some dispersive medium that affects the two wavefronts differently. These two wavefronts then pass through the second SHG, converting more of the fundamental into second harmonic. The fundamental is then filtered out, allowing the two second harmonic fields to produce an interference pattern whose fringe density is proportional to the dispersion of the fundamental and second harmonic wavefronts. The equation describing the fringes produced by the NLF is given by:

$$l\Delta n = m\lambda_s \quad (1)$$

Where l is the physical depth of the dispersive object, Δn is the difference in index between the fundamental and second harmonic wavefronts, m is the fringe order, and λ_s the second harmonic wavefront. It should be noted that this is the same equation for fringe shape for a Mach Zehnder with an equivalent wavelength of $\lambda_{eq} = \lambda_s (n_s - 1)/\Delta n$. Since Δn is typically on the order of 10^{-2} , λ_{eq} is about $50\lambda_s$. When the object is a reflecting surface immersed in a dispersive liquid, the NLF becomes analagous to the double immersion method in holography (Wyant 1978), in which the refractive medium surrounding an object is changed between exposures giving a fringe pattern with equivalent wavelength $\lambda_{eq} = \lambda/\Delta n$. The NLF reduces this procedure to one step, allowing dispersion to mimic the two different

Figure 3. Schematic of the NLF interferometer.

Figure 4. Photograph of the interferometer.

fluids. This allows the NLF to make real time measurements. The desensitization of this interferometer by the inverse of the dispersion allow us to study objects whose optical depth varies by much more than λ_s . I see the NLF as a stepping stone between common visible interferometers which contour wavefronts on the order of λ_s and Moire technique which can contour very large phase distortions.

If the fundamental wavefront is not planar to within the limits described by the crystal's critical angle, one must use NCPM crystals as the SHGs to prevent Maker fringes from being produced in one or both of the doublers. Since the variety of NCPM crystals at common laser wavelengths is limited, one of the goals for the materials search is to find a Type I NCPM for the fundamental wavelength of 1.32 μm . for use in future interferometers.

Consider what happens when a wedge of material with homogenous index is placed in the NLF (Figure 3). The length l of the wedge is given by:

$$l(x) = x \tan \alpha \quad (2)$$

Where α is the wedge angle and x is the distance down the wedge. With this l we can now write the equation of the fringes produced.

$$m\lambda_s = x \tan \alpha (n_s - n_f) \quad (3)$$

This is an equation for straight equally spaced fringes perpendicular to the x direction with fringe spacing Δx given by:

$$\Delta x = \lambda_s (\tan \alpha (n_s - n_f)) \quad (4)$$

If the wedge is a birefringent, nonlinear material transparent to both the and fundamental and second harmonic, and care is taken to excite

the proper polarizations, knowledge of the wedge angle allows us to extract directly the quantity $n_s - n_f$. This quantity is of interest in determining phase matching angles or temperatures in a search for new nonlinear materials. Hence the NLF directly measures the linear optical properties involved in phase matched SHG.

In a Type I configuration the index condition is given by:

$$n_{of} = n_{es} \quad (5)$$

If a wedge of a Type I phase matched material is placed in the NLF the fringe density approaches zero. This is because both fundamental and second harmonic refract through the wedge at the same angle. When this happens we should just see a spot of second harmonic with no fringe structure. When this crystal is not phase matched the fundamental and second harmonic do not refract equally, causing the two red wavefronts to be separated in angle causing straight, equally spaced interference fringes to appear.

In a Type II configuration there are two possible ways to measure phase mismatch. In the first technique one compares each of the two fundamentals with the one second harmonic, one pair at a time. Since the Type II phase matching condition reads:

$$\frac{n_{of} + n_{ef}}{2} = n_s \quad (6)$$

Type II phase matching through a wedge of the material being tested is achieved when the second harmonic refracts between the two fundamentals (Figure 5). When this wedge is placed in the NLF, phase match is observed when the fringe densities are equal for measurements made comparing the angular separation of each fundamental

Figure 5. Type II through a Wedge.

with the second harmonic. Phase mismatch is observed by noting different fringe densities for each of the two measurements. These measurements (both Type I and Type II) are made by placing the sample wedge between CPM SHGs using collimated laser beams. The Type II measurement can be made in one step if a Type II SHG is used as the second doubler. This technique combines both e and o fundamentals to produce a second harmonic that will then interfere with the second harmonic produced initially. Phase matching here is observed when the fringe density approaches zero, as in the Type I phase matching test. The main drawback to this approach is that if wedge angles are increased to increase the sensitivity of the measurement, the two fundamentals refract well separated. The phase matching in the second SHG then needs to be noncollinear (Figure 6). A Type II doubler designed for collinear phase matching cannot be phase matched. For this reason the first method described making two separate measurements will be used to make the Type II measurements.

Orthogonal Dispersion

While measuring the first sample, calcite, for a Type I angle tuned phase match, a method was devised that not only allows a visual interpretation of phase mismatch by looking at fringe tilt, but also calibrates the unknown dispersion of the sample wedge with the known dispersion of a reference wedge. Use of this method also enhances the sensitivity of measurement.

The technique is known as orthogonal dispersion because a wedge of known angle and dispersion is placed between the sample

Figure 6. Phase matching.

wedge and second doubler with it's wedge orthogonal to the sample wedge. Since the reference wedge is made of an isotropic material it can never be phase matched. The reference wedge alone produces straight, equally spaced fringes perpendicular to the variably spaced fringes produced by the sample wedge. These fringe patterns add to make the fringes tilt. The equation for the orientation of this combined fringe pattern is given by:

$$m\lambda = \tan \alpha_s (n_{ss} - n_{fs}) x + \tan \alpha_r (n_{sr} - n_{fr}) y \quad (7)$$

Here α_s, α_r are the sample and reference wedge angles respectively, $(n_{ss} - n_{fs})$ and $(n_{sr} - n_{fr})$ are the dispersion of the sample and reference wedge respectively, m is the fringe order, and λ_s the second harmonic wavelength. It is also assumed here that the sample wedge is opened or closed in the x direction, while the reference wedge is opened or closed in the y direction. The tilt changes as the dispersion in the sample wedge is varied. We can now solve for $n_{ss} - n_{fs}$, the quantity of interest, in terms of the known and measured quantities:

$$n_{ss} - n_{fs} = \frac{\tan \theta \tan \alpha_r (n_{sr} - n_{fr})}{\tan \alpha_s} \quad (8)$$

where θ is the angle measured from reference wedge fringes alone.

One nice feature of the technique of orthogonal dispersion is that one need not make these calculations while performing the experiment. Type I phase mismatch can be observed simply as a departure in angle of the fringes produced by combined sample and reference wedges from the fringes produced by the reference wedge alone. If the reference wedge produces horizontal fringes alone, we

manipulate the sample either in angle or temperature to try to get the combined fringes to be horizontal also. When this is observed, the sample is at, or very near phase match.

The Type II configuration is a bit more involved, but the technique of orthogonal dispersion still allows visual interpretation of the data. As described before, a Type II phase match is observed when the fringe densities obtained by individually interfering the second harmonic produced by the first doubler with the second harmonic produced by each of the two individual fundamental eigenvectors are equal. When an orthogonal wedge is placed between the sample and the second doubler, Type II phase match is visually observed when each fundamental "channel" produces equal and opposite fringe tilt relative to the fringes produced by the reference wedge alone.

Orthogonal dispersion measurements are particularly helpful in a search for NCPM materials. By measuring fringe angles at temperatures T_1, T_2 , we can predict phase matching temperatures even if phase match cannot be obtained in the experimental setup. This is a significant advance in NCPM materials studies as previous experiments on these temperature tuned materials were tedious, with very little insight gained if the phase matching temperature could not be obtained. As an example of how this might work consider a Type I NCPM configuration. Fringe angles θ_1 and θ_2 are measured at temperatures T_1 and T_2 . The difference in index is described by equations of the form of equation 8. Then:

$$\frac{\partial(n_s - n_f)}{\partial T} = \frac{n_s - n_f|_{T_1} - n_s - n_f|_{T_2}}{T_1 - T_2} \quad (9)$$

For an NCPM:

$$n_f|_{T_1} + \frac{\partial n_f}{\partial T} (T_{pm} - T_1) = n_s|_{T_1} + \frac{\partial n_s}{\partial T} (T_{pm} - T_1) \quad (10)$$

This equation can now be solved for the unknown phase match temperature, T_{pm} :

$$T_{pm} = \frac{n_s - n_f|_{T_1}}{\left[\frac{\partial(n_s - n_f)}{\partial T} \right]} + T_1 \quad (11)$$

Similar calculations can be done for the Type II NCPM case. They will be presented along with results for measurements of Gadolinium Molybdate in the materials section of this dissertation.

We will now examine the sensitivity of the technique of orthogonal dispersion for measuring index differences. Assuming well known reference wedge angles and indices (1° fused silica), known sample wedge angle (10°), and fringe angle resolution of plus or minus 1° , the typical sensitivity of this measuring technique near phase match is on the order of $\Delta n = 10^{-5}$. This sensitivity is much better than if one had to read changes in fringe density directly, since changes of fractions of a fringe are difficult to observe.

There are two other methods for making phase mismatch measurements. The first measures the angular separation of the eigenvectors by placing a lens after the sample wedge, and examining the separation of spots in the focal plane. This technique still requires the first doubler to supply both fundamental and second harmonic of the proper polarizations to the sample, however if the fundamental can be detected the second doubler can be eliminated. This technique's sensitivity near phase match is diffraction limited. To resolve peaks from two airy patterns, they must be separated by $2.44 \lambda f\#$. For typical wedge angles, focal lengths and beam diameters the sensitivity of this type of measurement is, at best, $\Delta n = 10^{-4}$. This calculation ignores any aberrations in the imaging system that will affect the Airy disc, as well as ignoring resolution limits on the imaging system. Examining fringe tilt using orthogonal dispersion increases the sensitivity of the phase mismatch measurement by at least one order of magnitude.

Another method of measuring phase mismatch has been suggested by G.R. Meridith of the Xerox Webster research labs in a private communication with M. Cervantes (Hopf and Cervantes, 1982). He suggests that if one use a very high power laser such that second harmonic light can be generated way off phase match, one can observe the Maker fringes generated by the sample to make an equivalent and somewhat easier measurement of Δn in nonlinear crystals. I see two problems with this technique. First is that one must have a very powerful laser. This type of laser does not exist at all wavelengths

of interest. The second problem is sample damage. At very high powers nonlinear materials can be permanently damaged, and even destroyed. The higher the nonlinearity the more likely the material is to be destroyed. This is because large nonlinearities usually mean large indices of refraction which indicate large electron cloud excursions. This means that the electrons can be ripped from their potential wells in the presence of intense fields. Some samples examined for this dissertation are extremely rare, so the possibility of bulk damage excludes this technique.

Spatial Correlations and Convolutions

Other nonlinear interferometers can be configured so that the second harmonic output is either a correlation or convolution of the two input waves involved in the nonlinear interaction. This type of interferometer is not common path, but could be useful in wavefront analyses, image recognition and possibly robot vision. Correlators of this kind are commonly used to measure extremely short optical pulselengths.

The key to this technique is the convolution theorem as described in Gaskill (1978). This theorem states that the Fourier transform of a product of two functions is the convolution of these functions. Since the second harmonic intensity is proportional to the product of the two input fields, taking the fourier transform optically by placing the image plane in the transform plane of a lens produces the correlation of the two fields. If the original object is axially symmetric one obtains the autoconvolution. The

autoconvolution process is extremely useful in image recognition (Gaskill 1978).

Type II doublers are a natural choice for nonlinear correlators since linearly polarized light is easily split into two orthogonal polarization components. One or both of these wavefronts can be encoded with phase information, then recombined in a collinear Type II phase match to produce a second harmonic signal that when inversely fourier transformed is the convolution of the two orthogonal polarizations. The apparatus for performing a Type II convolution is shown in Figure 7. The first lens takes the fourier transform of each of the orthogonally polarized images. The doubler is placed at this transform plane, producing a second harmonic signal proportional to the product of the transforms field distributions. At this point the fundamental wavelength can be filtered. The second lens takes the inverse Fourier transform of the second harmonic field intensity distribution, which is simply the convolution of the initial images at the fundamental wavelength.

One can also make a convolution using Type I doublers by using noncollinear phase matching (Figure 8). Part of the beam is split off encoded with phase information and then recombined with the rest of the beam inclined at some angle. These two fields are then individually tranformed by the first lens. A doubler designed for a noncollinear phase match is placed at this transform plane producing a

L = laser
 c = collimator
 pb = polarization beamsplitter
 m = mirror
 f = focal length
 F = infrared blocking filter

Figure 7. Type II all optical convolver.

L = laser
 c = collimator
 bs= beamsplitter
 m = mirror
 f = focal length
 F = infrared blocking filter

Figure 8. Type I all optical convolver.

second harmonic signal. By taking the inverse transform of the second harmonic signal, one again obtains the convolution of the two input fields.

The Type II configuration is preferable because it minimizes phase matching problems associated with noncollinear interactions. It would also be advantageous to use a Type II NCPM crystal because of the much larger critical angle obtained with this kind of phase match, but at present no such crystal is known to exist. We will examine a crystal, gadolinium molybdate (GMO) as a sample wedge in the NLF interferometer to try to see if this crystal is suitable for use as a Type II NCPM doubler for convolution applications at 1.3 μm .

Experimental Considerations

Most of the experiments measuring phase mismatch were performed at a fundamental wavelength of 1.318 μm . This wavelength promises to be of importance because of the high power available from the chemically pumped iodine laser now under development, and the lower power available from diode lasers used for communication through optical fibers because of low modal dispersion and losses at this approximate wavelength.

Our source of 1.318 μm . radiation is a Nd:YAG pulsed laser modified to work at the longer than usual wavelength by using an intracavity prism for wavelength selection. The laser is Q-switched electro-optically, generating a maximum of 13 millijoules in about 20 nanoseconds for a peak power of 850 Kwatts. This amount of power was rarely used in these experiments. The beam was usually expanded and

collimated using a Newtonian telescope with various magnifications. A pinhole spatial filter is made by placing a piece of aluminum foil in the focal plane of the first lens.

The reference harmonic generators are stock Inrad angle tuned Type I SHG's made of potassium dideuterium phosphate ($\text{KD}^* \text{P}$). They have a 13 mm diameter clear aperture, and a 15 mm. interaction length. These doublers produce visible red light from the infrared fundamental. They are in a fluid filled cell with the entrance window antireflection coated at $1.318 \mu\text{m}$. and the exit window antireflection coated at $.659 \mu\text{m}$. to prevent multiple reflections in the crystals. In their usual orientations these doublers produce vertically polarized second harmonic from the horizontally polarized infrared. If the SHG is tipped 45° we produce second harmonic polarized at 45° . We will use this feature to create polarizations that mimic a Type II configuration.

When a sample wedge of the nonlinear material of interest is placed between these two phase matched reference SHG's fringes will appear whose density is proportional to the phase mismatch. Placing an orthogonal reference wedge between the sample and second doubler allows us to interpret phase mismatch by examining fringe tilt. The wedges are oriented by looking at retroreflections from isotropic wedges using a helium neon laser. These retroreflections are either made vertical or horizontal depending on the desired orientation. The birefringent wedges were oriented by positioning the two spots transmitted by the crystal. The reference wedges used for most of

the experiments are 1 and 3 degree wedges made of fused silica. Fused silica was used because of the stability of the known indices of refraction for different lots of material. The accuracy of the wedge angle is claimed, by the manufacturer, to be plus or minus 1 arc minute. Another reference wedge was made with a larger wedge angle out of the same fused silica. Its wedge angle is $14.5^\circ \pm 2'$.

After the second doubler the infrared is blocked using a 3 mm thick piece of KG 4 heat absorbing filter glass made by Schott Glass Co. The imaging lens, as well as all the lenses used in these experiments are plano convex lenses made of crown glass and manufactured by Rolyn Optical. Interferograms are then recorded on film using a Pentax 35 mm camera back. Fringes were observed visually through the camera viewfinder, then subsequently recorded by allowing a few pulses to strike the film while the shutter remains open for a second or so. Neutral density filters were sometimes used when smaller beam diameters created higher power densities that would saturate the film.

CHAPTER 4

MATERIAL CONSIDERATIONS

The number of known phase matchable SHGs at the fundamental wavelength of $1.3\text{ }\mu\text{m}$ is limited. The research done for this dissertation not only introduces a technique general enough for measuring phase mismatch at many other wavelengths, but identifies some viable candidate materials for SHG at $1.3\text{ }\mu\text{m}$.

The measurements made do not concentrate on the magnitude of the material's quadratic nonlinearity, as many nonlinear optical materials studies have. A large nonlinearity is important for efficient SHG when the electric field strengths are well below the material's bulk damage threshold. However, as the field increases to near the damage threshold, a material with a large nonlinearity is no longer desirable. This is because materials with large nonlinearities usually experience large electron cloud movement from equilibrium, making it easier to ionize these materials with intense electric fields. In high power applications choosing a nonlinear material involves making tradeoffs between the size of the nonlinearity and the material's damage threshold.

Another important consideration in choosing a crystal for SHG is its optical quality. If the crystal distorts both the fundamental and second harmonic phasefronts, any advantage that would have been gained by switching to a shorter wavelength to decrease the effects

Table 1. Indices of refraction at wavelengths of interest.

1.064 μm UNIAXIALS

	n_{of}	n_{os}	n_{ef}	n_{es}
CALCITE	1.6432	1.6631	1.4796	1.4885
LITHIUM NIOBATE 25°C	2.2321	2.3232	2.1560	2.2342

BIAXIALS

	n_{of}	n_{bf}	n_{yf}	n_{os}	n_{bs}	n_{ys}
GMO	1.8141	1.8145	1.8637	1.8549	1.8553	1.9103
TMO	1.8222	1.8226	1.8704	1.8646	1.8650	1.9186
KTP	1.7402	1.7469	1.8304	1.7789	1.7924	1.8873
LITHIUM SULFATE	1.4534	1.4677	1.4771	1.4638	1.4789	1.4883

1.318 μm UNIAXIALS

	n_{of}	n_{os}	n_{ef}	n_{es}
CALCITE	1.6391	1.6546	1.4782	1.4846
LITHIUM NIOBATE 25°C	2.2197	2.2797	2.1453	2.1970

BIAXIALS

	n_{of}	n_{bf}	n_{yf}	n_{os}	n_{bs}	n_{ys}
GMO	1.8096	1.8100	1.8586	1.8354	1.8358	1.8880
TMO	1.8175	1.8179	1.8652	1.8443	1.8447	1.8955
KTP	1.7342	1.7399	1.8224	1.7608	1.7707	1.8598
LITHIUM SULFATE	1.4508	1.4644	1.4737	1.4595	1.4745	1.4842

of diffraction will be lost. Since, as previously mentioned, the size of the nonlinearity is not exclusively important, future research into nonlinear materials will have to focus more on the optical quality of the materials, as well as the bulk damage thresholds to fully take advantage of the shorter wavelengths produced by the SHGs. Interferograms made using the NLF of various materials to measure phase mismatch give us an indication of the crystal's optical quality. In spite of this, interferograms can be made with materials so poor that it is improbable that any other method could so precisely analyze the linear optical properties (the method of minimum dispersion is usually used to measure indices for phase match calculations).

In this chapter we describe materials and measurements made on these materials in a search for SHGs at 1.3 μm . The first experiments were performed on a piece of calcite. Although calcite's crystal symmetries prohibit efficient second harmonic due to a dipolar nonlinearity, quadrupolar SHG can be produced if one can phase match the interaction. Calcite is an inexpensive sample with large birefringence, allowing us to test the technique without risking rarer samples. Measurements were then taken from lithium niobate for a Type I NCPM, gadolinium molybdate for a Type II NCPM, and KTP for a Type II angle tuned phase match. Lithium sulfate is suggested for a Type I NCPM. The indices of materials at wavelengths of interest examined for this dissertation can be found in table 1. The properties of these materials as well as test results will be presented in this chapter.

Calcite

Calcite (CaCO_3) crystals are commonly manufactured into polarizers for laser applications (Machewirth 1979). Calcite crystals are available in large rhombs, however, one must be careful to avoid samples with visible striae that will scatter light. Calcite belongs to the crystal class $\bar{3}m$; it is a trigonal crystal having a center of inversion symmetry. This symmetry does not allow dipolar SHG, as the second order coefficient of the polarization must be zero. Thus the nonlinear effects observed in calcite are of one order higher nonlinearity than effects observed in noncentrosymmetric crystals. This means that the magnitude of the effect is down by a factor of about a thousand from the SHG produced by noncentrosymmetric crystals. In calcite there is an induced quadrupole moment proportional to the square of the applied field. The quadrupole expansion is commonly written:

$$Q_{ij} = \eta_{ijkl} E_k E_l \quad (1)$$

The effects of this induced quadrupole are combined with higher order dipolar terms to produce an effective quadrupole moment at the second harmonic frequency. This is known as quadrupole second harmonic generation (QSHG). Additional second harmonic can be produced by applying a DC electric field to the calcite crystal. This field upsets the center of symmetry of the crystal allowing dipolar SHG to occur. This is electric field induced SHG or ESHG. Further reference on how this quadrupole radiates can be found in Bjorkholm and Siegman (1967) and Hopf and Stegeman (to be published).

Optically, calcite is a negative uniaxial crystal. This means that the ordinary index of refraction is higher than the extraordinary index. The ordinary axis transmits light over the spectral range of 0.26 to 1.75 μm , while the extraordinary axis transmits light over the broader range of 0.214 to 3.3 μm , with a slight dip at 2.5 μm . This property is known as dichroism, and is due to the anisotropic nature of its molecular structure (Figure 9a). Calcite has a perfect cleavage plane parallel with the rhomb face, with the extraordinary (optic axis) at an angle of $44^\circ 37'$ (Figure 9b). The dispersion curves of calcite as well as the phase matching configuration are shown in Figure 10. These curves were plotted by an IBM personal computer after using a program written by Mike Nofziger of the Optical Sciences Center (OSC) to fit dispersion curves to known data points using a Herzberger fit. The data points were obtained from the Infrared Handbook by Wolfe (1975). The formulae for these curve fittings can be found in the computer program in Appendix A.

In order to observe QSHG from calcite we will use a Type I angle matched configuration (Figure 11). Terhune et al.(1962), have also observed somewhat weaker QSHG from a Type II configuration. In our configuration, the fundamental is polarized along the o axis (perpendicular to the optic axis), with the second harmonic polarized perpendicular to the fundamental. The crystal is then angle tuned between the o and e axes to index match the fundamental and second harmonic. Measurements were taken from calcite at both 1.06 and 1.32 μm . The calcite crystal was cut and polished so that phase matching

Figure 9a. Calcite's molecular structure.

Figure 9b. Optical axis configuration in calcite.

Figure 10. Phase matching considerations in Calcite.

a. Indices of refraction.

b. Type I angle tuning at 1.32 μm .

Figure 11. Type I angle tuned phase matching at 1.32 μm . in Calcite.

oa=optic axis; o=ordinary axis

could occur normal to this face at 1.06 μm . The opposite face was cut for phase matching normal to this face at 1.32 μm ., forming a wedge whose angle is the difference between the phase match angles at these two wavelengths. The index matching condition for these two cuts is given by:

$$n_{\text{of}} = n_{\text{ef}}(\theta) \quad (2)$$

θ_{pm} , the phase matching angle is calculated to be 18.5° and 16.3° respectively for 1.06 and 1.32 μm . The angles are measured from the optic axis. If the crystal is cut to these angles a wedge of 2.2° is generated. The walkoff of the second harmonic relative to the fundamental is calculated to be 3.7° at 1.32 μm .

At 1.06 the calcite crystal was placed between a lithium niobate crystal configured for a NCPM and a lithium iodate crystal that is critically phase matched. Although another NCPM niobate crystal was available for use as the second doubler, results showed that using a CPM crystal prevented the production of extraneous fringe patterns due to multiple reflections in the calcite that would be phase matched had a NCPM crystal been used. The calcite crystal was mounted onto the 3° orthogonal reference wedge with wax. Ethylene glycol, an index matching fluid with index of 1.434 was placed in the calcite-reference wedge interface, reducing reflections from both the exit face of the calcite and entrance face of the reference wedge. As described in ch. 3 phase match is observed by interpreting fringe tilt as the crystal is rotated about a horizontal axis. These results are shown in figure 12. Note that the reference wedge alone produces

Figure 12. NLF interferograms of calcite at 1.06/.53 μm .

vertical fringes. This is slightly different from the analysis in ch. 3, where the reference wedge produced horizontal fringes.

Phase matched SHG was not observed visually with the power levels available to us using our 1.06 μm . laser which has a modest peak power of 250 Kwatts. SHG was observed visually using ≈ 30 Mwatts from a collimated beam with a diameter of ≈ 1 cm from a different YAG laser at 1.06 μm . This SHG was observed within the experimental uncertainty from the angular position predicted by the interferometric phase match predictions and within a degree of the face normal.

The experiments were then repeated at 1.32 μm ., by simply remounting the crystal, flipping entrance for exit faces. These results are shown in Figure 13. To check the degree of accuracy with which we can compare the experimentally derived Δn with the Δn predicted by available references we must first solve the problem of coupling Snell's law of refraction with the angle tuned index. The index that the second harmonic sees is dependant on the angle of the k vector with respect to the optic axis. This angle in turn is dependant on Snell's law, which depends on the index seen by the second harmonic. This problem is solved by making iterative approximations. This approximation begins by solving for $n(\theta)$ at normal incidence. This calculated index is then used to calculate the refraction using Snell's law at intervals of 1° . This angle is then added or subtracted from the initial angle from the optic axis to the surface normal, which then generates a new index, which is then used in Snell's law for the next iterative step of rotating the crystal 1° .

Figure 13. NLF interferograms of calcite at 1.3/.66 μm .

These generated indices for both 1.06 and 1.32 μm . are shown in Figure 14, along with the predicted Δn as a function of the angle of incidence. These figures are then compared with the experimentally derived Δn in intervals of 3°.

Lithium Niobate

Lithium niobate (LiNbO_3) is one of the most versatile materials used in photonics applications. It is commonly used for nonlinear optical, electrooptic, acoustooptic, and pyroelectric applications. High optical quality crystals are synthetically grown using the Czochralski pulling technique from melts with various compositions. Niobate crystals are not hygroscopic, that is, they do not dissolve in the presence of water. This is a significant advantage over other standard nonlinear optical crystals such as KDP. Another advantage of niobate is that it is easily fabricated and polished by standard glass working techniques. Niobate crystals are also mechanically stable, they are not adversely affected by thermal or mechanical shock. More importantly, lithium niobate has a large nonlinear coefficient, making it an ideal material for low to medium power nonlinear frequency conversion. Bulk damage is observed at power levels on the order of megawatts per square centimeter, but more important is the refractive index damage observed in the presence of moderate power levels in the visible spectrum. This damage can be removed by heating the crystal above 160° C, but since one may need to operate below this temperature for NCPM SHG applications at fundamental wavelengths below 1.1 μm ., doping the niobate with magnesium oxide (MgO) in the

Figure 14. Calcite results (Δn) experimental vs. (Δn) theoretical.

proper quantity has shown to all but eliminate the optical damage caused the generated visible light. The most interesting feature of lithium niobate for use in NCPM applications is the temperature sensitivity of the birefringence, allowing these crystals to be phase matched over an extraordinary range of fundamental wavelengths. If one restricts the temperature range between $0^{\circ} - 300^{\circ}$ C one can produce second harmonic in a NCPM configuration throughout most of the visible spectrum (green to red).

Lithium niobate is a negative uniaxial crystal with a rhombohedral structure, belonging to the crystal class 3m. It is ferroelectric. A crystal is said to be ferroelectric if the odd rank tensor coefficients, beginning with the linear polarization can be changed in the presence of an external electric field. The problem with ferroelectrics is that they commonly have a domain structure that shows up optically as a region of strain or refractive index inhomogeneity that results in excessive scattering and phase distortion, as well as impeding energy flow. Lithium niobate originally exhibited a domain structure. Lithium niobate can be made single domain during the growth process by passing a small poling current throughout the pull rod. The act of poling aligns the ferroelectric domains in the same direction. Poling is achieved by holding the crystal at a sufficiently high temperature for the domain structure to be reversible. The applied electric field aligns the domains in a single direction. When the crystal is cooled the domain structure is no longer reversible, so when the electric field is removed, the crystal remains single domain.

In initial experiments with lithium niobate Byer et al.(1970), it was discovered that the refractive indices varied from boule to boule, with the birefringence also varying spatially. Nonuniform birefringence drastically affects the efficiency of phase matched interactions, since different points in the crystal have different phase match conditions. It has since been shown that compositional changes are responsible for both variations in index, and also for the birefringence variations. High quality crystals can be obtained only if growth conditions and melt compositions can be carefully controlled. Byer et al. (1970) found that a certain composition, known as congruent, can eliminate variations in index for different boules, as well as having virtually no birefringence variations. Congruent lithium niobate has a composition that corresponds to a mole ratio of $\text{LiO}_2 / (\text{LiO}_2 + \text{Nb}_2\text{O}_5) = 0.486$ rather than the 0.5 ratio of stoichiometric lithium niobate. Lithium niobate samples used in experiments measuring phase mismatch were grown from a congruent melt by Crystal Technology Inc.

The indices of refraction of congruently melted lithium niobate were obtained from a paper by Nelson et al. (1974), then fitted to a Herzberger curve using the same program used to fit the other material's dispersion curves. The Herzberger fit is then used to plot the dispersion curves of the ordinary and extraordinary axes Figure 15, as well as generate indices at wavelengths of interest table 1. Lithium niobate also has an intensity dependant refractive

Figure 15. Phase matching considerations in Lithium Niobate.

a. Indices of refraction.

b. Type I 90° phase matching at 1.32 μ m.

index. This means that the refractive index can be written as:

$$n = n_0 + n_2 I \quad (3)$$

where n_0 is the refractive index measured at low irradiances. The value of n_2 has been measured by Lattes, et al. (1983) to be approximately $3 \times 10^{-9} \frac{\text{cm}^2}{\text{Mw}}$. Since we are using about 0.5 Mw in a 5 mm diameter spot, the intensity dependant index is negligible, and hence will be ignored. Although negligible in our case, lithium niobate waveguides can enhance this effect by confining the electric field to extremely small dimensions, allowing lithium niobate to be used as an intensity dependant switch (Sarid and Sargent 1982).

Lithium niobate will be tested for a Type I NCPM. The fundamental is polarized along an o axis, with the second harmonic polarized along the optic axis, forming an ooe configuration (Fig. 16). At room temperature one can see that the phase matching condition is far from being satisfied. However, in lithium niobate the extraordinary index changes so drastically with temperature that by heating the crystal to about 300° C phase matching can be achieved. Temperature tuning data given by Byer et al.(1970) shows that the temperature tunability of lithium niobate changes with pump wavelength. Experiments performed at 1.32 μm . will measure the temperature tuning coefficients involved in SHG at this wavelength. This result is not only important for predicting phase matching temperatures, but also gives an indication on the temperature tolerances for holding this phase match.

Figure 16. Type I 90° phase matching at 1.32 μm . in Lithium Niobate.

o=ordinary axis; e=extraordinary axis

In a crystal with 3m symmetry the effective coefficient of the nonlinearity is given by

$$d_{\text{eff}} = d_{15} \sin \theta + d_{22} \cos \theta \quad (4)$$

Where θ is the angle between the k vector and the optic axis. For an NCPM configuration $\theta = 90^\circ$ so $d_{\text{eff}} = d_{15}$. The value of this coefficient is given by Miller et al.(1966) to be

$$d_{\text{eff}} = 5.82 \pm 0.85 \times 10^{-12} \text{ m/v} \quad (5)$$

This large nonlinearity is consistent with lithium niobate's large refractive index, which exceeds 2.2.

Two sample wedges were cut and polished by the O.S.C.'s optics shop, from a slice cut off a Z axis grown boule of lithium niobate supplied by Crystal Technology Inc. The first wedge has a modest wedge angle of 5° , while the second wedge has a more severe wedge angle of 20.25° . These two samples were measured for phase mismatch as the crystal is scanned between room temperature, 25°C , and 320°C . Different combinations of sample wedge angle and orthogonal reference wedge angles are made to examine how the sensitivity of the measurements change as these parameters are adjusted.

The most sensitive measurements are made by using the larger wedged sample with the reference wedge with the smallest angle. In our most sensitive measurement we compare the 20° sample wedge with the 3° reference wedge. These measurements were made with a 3 mm. collimated laser beam at $1.3 \mu\text{m}$. The increase of sensitivity is only experienced near phase matching. Fringes observed near room temperature are very tightly packed, and may not be resolved after

reproduction. As the temperature is increased from room temperature, one observes the fringe density decrease but the fringes do not start tilting until about 200° C. If one is to use fringe tilt to extract the temperature tuning coefficients, as described in chapter 3, the range of temperature is reduced for this combination of wedges. Interferograms made comparing the large niobate wedge with the 3° orthogonal reference wedge are shown in Figure 17. Note how the fringe tilt changes drastically with temperature near phase match, and the ellipticity of the beam caused by the extreme wedge angle. Phase matching is indicated by horizontal fringes. Since the sample is producing its own second harmonic, one should note the decrease in fringe visibility caused by the addition of a third field in the interference pattern, as well as the extra laser speckle sprayed around the edge of the interferograms at phase match. Phase matching occurred at $295^{\circ} \pm 2^{\circ}$ C. The temperature tuning coefficient can be extracted from these interferograms and is calculated to be $6.9 \pm 2.7 \times 10^{-5} \frac{1}{C}$ for this combination of wedges.

Interferograms comparing the sample wedge with both the 1° and 3° reference wedges are shown in Figs. 18 and 19 respectively. The diameter of the beam was increased from 3 to 5 mm. for these tests to observe more fringes across the field. As the temperature is increased, one observes the fringes tilting more towards horizontal, as well as decreasing in density. Any curvature in the fringes is caused by thermal gradients in the niobate. Note that the fringes on either side of phase match tilt more using the smaller reference

Figure 17. NLF interferograms of 20° lithium niobate wedge with 3° reference wedge.

Figure 18. NLF interferograms of 5° lithium niobate wedge with 1° reference wedge.

Figure 19. NLF interferograms of 5° lithium niobate wedge with 3° reference wedge.

wedge, indicating an increase in sensitivity due to the decrease in the reference wedge angle. The temperature tuning coefficient is calculated to be $5.6 \pm 3.1 \times 10^{-5} \frac{1}{C^\circ}$ for the 1° reference test, and $6.9 \pm 1.6 \times 10^{-5} \frac{1}{C^\circ}$ for the 3° reference test. These data were obtained by a linear curve fits of (Δn) versus T from plots similar to Figure 20. This compares with $\frac{\partial(\Delta n)}{\partial T} = 8 \times 10^{-5} \frac{1}{C^\circ}$ as measured by Byer (1970) at the fundamental wavelength of $1.15 \mu m$.

Figure 20. Lithium niobate experimental results.

(3° reference wedge, 5° sample wedge)

KTP

During the last few years, a new nonlinear optical material has been developed that has become the frequency doubling material of choice for commercially built YAG laser systems. This crystal is KTP (KTiOPO₄). KTP has an extremely high bulk damage threshold, making it is useful for high power applications. KDP type crystals, currently in use for these applications, have relatively low nonlinear coefficients, stringent phase matching requirements, are hygroscopic, and do not yield high conversion efficiencies at power densities available from commercially built laser systems.

Originally developed by DuPont and now grown by Airtron, KTP crystals have a large nonlinear coefficient in addition to the high damage thresholds. The material has wide angular and temperature phase matching bandwidths and can be phase matched in either a Type I or Type II configuration. Furthermore, the material is nonhygroscopic, thermally and chemically stable. Except for the fact that crystals can only be grown up to about 5x5x5 mm., KTP is perhaps the most attractive nonlinear material available for high power, high conversion efficiency applications. A maximum energy conversion efficiency of 60 was observed at an incident intensity of 250 Mwatt/cm². Bulk damage is observed at a power density of about 400 Mwatt/cm² (Liu et al., 1982). Surface damage is observed well below this power density if the crystal has any dust or oil on the surface. We have found, however, some index inhomogeneities in the samples supplied by Airtron that cause some strange effects to be seen. These will be described in detail later in this section.

KTP has an orthorhombic structure and is of the point group $mm2$. It is biaxial, so there are three branches to the dispersion curve. The material should be ferroelectric because of symmetry conditions, suggesting the possibility of a domain structure. Standard methods for detecting domains such as viewing between crossed polarizers, electrostatic decoration of surface charge, scanning electron microscopy, and chemical etching failed to reveal any domain structure (Bindloss et al. 1976), yet this author believes some sort of domain structure exists because of the strange behavior observed in our samples.

KTP is grown using a hydrothermal technique, producing crystals that are water insensitive, easily cut and polished, and free from any visible strata that might scatter light. KTP is transparent from 0.35 to 4.5 μm . The lack of any color and the constant transmission throughout the visible and near IR, indicate that absorption is very small in this region.

The dispersion curves fig 21a, were plotted from the Sellmeier equation fitted to known index data points. The coefficients for this fit were obtained verbally from Dave Dentz of Airtron. They were then plotted using the graphics program listed in Appendix A.

For SHG at the fundamental wavelength of 1.3 μm ., a sample was obtained from Airtron that was cut for a Type II angle tuned phase match. KTP can be angle tuned in the YZ, XZ, or XY planes, however to reduce the amount of material used, the YZ configuration

Figure 21. Phase matching considerations in KTP at 1.32 μ m.
 a. Indices of refraction
 b. Type II angle tuned phase matching at 1.32 μ m.

was chosen. The phase matching configuration used is shown in figures 21b and 22. The sample obtained was cut for phase matching normal to the entrance face, with a 4° wedge at the exit face. The aperture size is approximately 3×3 mm., with an interaction length of 1.5 mm. In this configuration one fundamental along with the second harmonic are polarized along the X axis, and are o rays, while the other fundamental is an e ray that is tuned between the Y and Z axis such that the average index of the two fundamentals is equal to the index of the second harmonic. The angle calculated for this phase match is 39.9° from the Y axis. The relevant d_{eff} for this configuration is $d_{31} \sin \theta_{\text{pm}}$. Since $d_{31} = 6.5 \times 10^{-12}$ M/V for KTP,

$$d_{\text{eff}} = 4.2 \text{ M/V. (KTP)} \quad (6)$$

The calculated walkoff angle of the e ray fundamental is 2.6° . Intense phase matched second harmonic was observed for this configuration, however, when this crystal was placed in the NLF for phase mismatch measurements an odd behavior was observed.

It was first noted that when a circularly symmetric YAG laser pulse propagated through the crystal, a fringe pattern emerged. Further examination showed that each polarization produced it's own fringe pattern. When the KTP was phase matched, the second harmonic emerged in a fringe pattern. A representation of these fringe patterns are shown in Figure 23b. To try to understand what was happening, we placed a 300 mm. focal length lens immediately behind the crystal and looked at spots produced in the focal plane. To our surprise, 4 infrared spots and two visible spots were observed in a

Figure 22. Type II angle tuned phase matching at 1.32 μm . in KTP.

x, y, z = principle axes.

Figure 23. Observed behavior in KTP.

- (a) Input spot
- (b) Near field
- (c) Focal plane

pattern represented by figure 23c. It was found that each polarization was producing two spots. This is consistent with the observed fringe patterns, but is inconsistent with the common laws of crystal optics. This behavior had been previously not been observed, since most of this material is fabricated into cubes. What had been noticed, only in certain lots of material, is a double peak in the second harmonic produced as the crystal is tuned through phase match. Gary Vallaincourt, an employee of Quantronix Corp., a YAG laser manufacturer, recently reported to this author that some cubes of KTP had this double peak behavior. When KTP is phase matched, one can observe the second harmonic being produced inside the crystal. In crystals that showed the double peak behavior, Gary noticed gaps in the visible light inside the crystal, that would interchange bright for dark as the crystal is tuned to the other peak. It seems as if there is some sort of defect in some crystals of KTP that cause different parts of the crystal to have different orientations. The physical basis for this phenomenon is not well understood. Further research on this odd crystal behavior, probably starting with X-ray diffraction, will possibly explain this behavior. Because of this behavior, interferograms measuring phase mismatch could not be made.

Rare Earth Molybdates

Certain rare earth molybdates, in particular, gadolinium molybdate (GMO), with composition $Gd_2(MoO_4)_3$, , terbium molybdate (TMO) with composition $Tb_2(MoO_4)_3$, , and hybrids of terbium-gadolinium molybdates (GTMO) with compositions $Gd_x Tb_{2-x}(MoO_4)_3$, have dispersion

curves that are very close to exhibiting a Type II NCPM at a fundamental wavelength of $1.3 \mu\text{m}$. They have nonlinear matrix elements that allow the production of second harmonic, and are transparent from 0.4 to $4.5 \mu\text{m}$. If Type II phase matched second harmonic could be produced, these crystals could be used for optical convolvers, as described in chapter 3. Interferograms measuring phase mismatch were made from gadolinium molybdate, but extremely high scattering and wavefront distortion was observed. This scattering is believed to be caused by domain walls and small inclusions.

Rare earth molybdates are ferroelectric - ferroelastic crystals. This means that there is an interaction between an applied electric field and or mechanical stress field with the crystal's observed properties. At temperatures less than about 160°C the rare earth molybdates have an orthorhombic crystal structure, and belong to the point group $mm2$. At high temperature, the structure is tetragonal, with point group $42m$. When cooled or heated through this phase transition the equivalent $[110]$ and $[\bar{1}\bar{1}0]$ axes of the tetragonal

phase become the X and Y axes of the orthorhombic phase. Since these two orthorhombic axes are unequal, the crystal changes shape by a small but significant amount. In the orthorhombic state, polydomain twinning is often observed. Domain walls can be moved or temporarily eliminated by introducing a electric field or mechanical stress to the crystal. This is a direct consequence of the ferroelectric -

ferroelastic nature of these crystals. Zigzag domain walls were observed in the sample of gadolinium molybdate by simply illuminating the crystal with polarized light from a HeNe laser. Figure 24a shows the domains as seen in transmission through the crystal, while Figure 24b shows the high scattering caused by both domain walls and small inclusions. These inclusions were observed by placing a thin section ($\approx 50 \mu\text{m}$.) under a polarizing microscope. Domain walls must be eliminated if these rare earth molybdates are to become useful in nonlinear optical applications. However, this domain wall behavior, and its dependence on electric and stress fields, has attracted interest in device applications that in some way involve domain wall movement to modulate electromagnetic or acoustic energy (Kumada et al 1971).

Below the phase transition temperature the rare earth molybdates are optically biaxial, although only slightly. The indices of the a and b axes are so close that at first these crystals were mistaken for uniaxial crystals. As shown in Figs. 25 and 26, the dispersion curves of the rare earth molybdates can be altered by varying the composition. It should be noted that the separation of X and Y indices is exaggerated, as the real curves overlapped. Some composition will exhibit a Type II phase match near room temperature. The phase match configuration to be tested is shown in Figure 27. The relevant d_{eff} for this ooe configuration is d_{32} . The effective nonlinearities for GMO and TMO are given by

$$d_{\text{eff}} = 2.75 \times 10^{-12} \text{ M/V} \quad (\text{GMO}) \quad (7)$$

(a)

(b)

Figure 24. Domains in GMO.

- (a) Zigzag domains
- (b) Scattering laser light

Figure 25. Phase matching considerations in GMO.

a. Indices of refraction.

b. Type II 90° phase matching at 1.32 μ m.

Figure 26. Phase matching considerations in TMO.

a. Indices of refraction.

b. Type II 90° phase matching at 1.32μm.

Figure 27. Type II 90° phase matching at $1.32 \mu\text{m}$. in GMO, TMO.

x, y, z = principle axes.

$$d_{\text{eff}} = 2.52 \times 10^{-12} \text{ M/V} \quad (\text{TMO}) \quad (8)$$

Since the X and Y indices are so close, interchange of X and Y axes still allow a possible Type II phase match. In that case $d_{\text{eff}} = d_{31}$, which has an opposite sign, and is slightly larger. (Miller et al. 1970)

Since our laser is horizontally polarized, both crystals and orthogonal reference wedges were tipped at 45° , to excite both fundamental polarizations in the crystal. In addition, the first $\text{KD}^* \text{P}$ doubler used in the NLF was also tipped at 45° to excite only one red polarization. This also reduces the amount of red produced, increasing the contrast of the observed fringe patterns. The second doubler was not tipped so that either polarization could produce second harmonic in the second crystal. Since the two fundamentals emerge from the crystal at different angles, two sets of fringes are obtained, one at a time. One can observe the dispersion between either of the fundamentals and the one red polarization by simply angle tuning the second doubler to phase match one of the two fundamentals. The red produced by either of the fundamentals then interferes with the red produced by the first doubler, producing a fringe pattern whose density is proportional to Δn . When an orthogonal wedge is placed between the molybdate sample and the second $\text{KD}^* \text{P}$ doubler, Type II phase match is observed when equal fringe tilt is obtained, symmetric about the reference fringe tilt angle.

Samples were obtained from DuPont Inc. of various compositions. The sample chosen for testing was a piece of GMO with a

cleavage plane normal to the Z axis. It was not verified whether the X or Y axis was in the propagation direction, but since both orientations could possibly be phase matched, this was not of concern. Interferograms measuring Type II phase mismatch were made for GMO in spite of the poor optical quality of this crystal caused by polydomain twinning and scattering from inclusions. The fringe density shown in Figure 28 for the sample wedge alone is higher than those taken for both polarizations. This is because the film plane was moved further back, with a part of the interference field blocked by an aperture. This improved the quality of the interferograms obtained, without losing any fringe tilt information. Measurements showed that fringe tilt did not change for comparing the dispersion of the visible and infrared polarized along the same axis. There was, however, a small change of approximately 1° in fringe tilt observed while comparing the dispersion of the visible polarized along the X axis with the infrared polarized along the Z axis. Using the change in tilt to calculate the temperature tuning coefficient we find $(\partial(n_f - n_s))/\partial T \leq 2.7 \times 10^{-5} \frac{1}{C^\circ}$. Published results indicate this quantity to be $= 2 \times 10^{-5} \frac{1}{C^\circ}$ (Kojima and Nakamura 1978), consistent with our findings. To predict the Type II phase matching temperature we first write the phase matching condition;

$$n_{sY} = \frac{n_{fY} + n_{fZ}}{2} \quad (9)$$

If we now allow the indices to vary with temperature and make the approximation that $\partial n_{sY}/\partial T = \partial n_{fY}/\partial T = \partial n_Y/\partial T$, we find;

Figure 28. NLF interferograms of GMC at 1.32 μm .

$$2n_{sY} - n_{fY} - n_{fZ} \Big|_{T=T_0} = \frac{3(n_{fZ} - n_Y)}{\partial T} \Delta T \quad (10)$$

Here ΔT is the $T_{pm} - T_0$. If ΔT is positive we must heat the crystal. If ΔT is negative we must cool the crystal to achieve phase matching. On the basis of our results obtained, the phase matching temperature cannot be reached at any reasonable temperature for this sample of GMO.

Lithium Sulfate

Lithium sulfate ($Li_2SO_4 \cdot H_2O$) is a crystal which is not normally considered to be a nonlinear material for SHG. The only reference this author could find describing SHG in lithium sulfate is in a paper written by Hobden (1967). From index data available in this paper it appears that lithium sulfate could exhibit a possible Type I NCPM at 1.32 μm . with an adjustment of crystal temperature.

Lithium Sulfate is optically biaxial. Index of refraction data points from Hobden's paper were fitted to a two term Sellmeier fit allowing the dispersion curves to be plotted versus wavelength. These dispersion relations are shown in the computer program listed in Appendix A, and are plotted in Figure 29. Lithium sulfate is a monoclinic crystal with point group symmetry 2. The phase matching configuration tested in lithium sulfate is a Type I NCPM, and is shown in Figure 30a. The orientation of eigenvectors is an ooe, with the horizontally polarized fundamental along the c axis and the vertically polarized second harmonic along the b axis as shown in figure 30b.

Figure 29. Phase matching considerations in Lithium Sulfate.

a. Indices of refraction.

b. Type I 90° phase matching at 1.32 μ m.

Figure 30. Type I 90° phase matching at 1.32 μm .
in Lithium Sulfate.
 a, b, c = principle axes.

The relevant d_{eff} for this configuration is given by d_{22} . It has a value of

$$d_{\text{eff}} = .33 \times 10^{-12} \text{ M/V} \quad (11)$$

The magnitude of this nonlinearity is a factor of ten less than the nonlinearities in more common SHGs. The size of the nonlinearity in lithium sulfate is consistent with the relatively small indices of less than 1.5. This low index hints that lithium sulfate might have a large damage threshold, but no data was available that could verify this.

Another problem with lithium sulfate is that since water is part of its chemical composition, it is hygroscopic. Combining this with the fact that the crystal needs to be cooled below the dew point means that a protective cell must be configured. The good news is that there are index matching fluids that exist at the low indices of lithium sulfate that could reduce reflections at the cell window - crystal interface.

If one were to test lithium sulfate for a Type I NCPM another advantage of our testing technique would become apparent. Although it was believed that cooling of the crystal was needed to phase match, we do not know how much cooling was necessary. If the only temperature controller available for these experiments was a crystal oven, our testing technique allows us to predict the phase match temperature, even if we changes the temperature in the wrong direction. We also would not need to build a crystal housing to prevent water vapor from condensing on the hygroscopic crystal.

Lithium sulfate crystals were not available at the time of the experiments to test.

CHAPTER 5

CONCLUSION

The nonlinear Fizeau interferometer has proven itself to be a powerful tool for a nonlinear optical materials search. Measurement of phase mismatch in possible SHGs is feasible, and easily accomplished using the NLF interferometer. Visual interpretation of the data via orthogonal dispersion is the key to this measuring technique, however, useful information can alternatively be extracted by accurately analyzing fringe tilt from photographs of the fringe patterns.

The major problem with analyzing NCPM materials was thermal gradients. These gradients could not be avoided in our experimental setup because our crystal oven had only two settings, on and off. Since temperature had to be scanned, thermal gradients in our sample of lithium niobate caused fringe tilt to vary greatly in one exposure. This made accurate temperature tuning data difficult to extract from the interferograms, and accounts for the large error bars obtained in figure 23. To extract this temperature tuning coefficients accurately one would need more accurate temperature control.

In the case of measuring Type II phase match in GMO, which has a smaller temperature tuning coefficient than lithium niobate, we did not see these thermal gradient effects. Interferograms made and analyzed for GMO agreed with the index differences that have been

previously published, yet phase matching cannot be achieved at any reasonable temperature. This along with domain problems, make this GMO sample unsuitable for a SHG at $1.32\text{ }\mu\text{m}$. Another composition of rare earth molybdate might work since the GMO sample is so close to the Type II phase match condition.

KTP can be phase matched to produce intense second harmonic at $1.32\text{ }\mu\text{m}$., although some strange crystal behavior prohibited production of NLF interferograms.

Calcite crystals were also tracked through phase match by placing them in the NLF, allowing us to accurately predict the angle at which quadrupolar second harmonic could be observed.

APPENDIX A

DISPERSION CURVES

(Written for IBM personal computer)

```

10 SCREEN 0
20 KEY OFF
21 PRINT "1. SUCROSE"
22 PRINT "2. NIOBATE"
23 PRINT "3. GMO"
24 PRINT "4. TMO"
25 PRINT "5. CALCITE"
26 PRINT "6. KTP"
28 PRINT "7. LITHIUM SULFATE": PRINT "99. END"
29 INPUT "FORMULA TO USE" ;N: IF N=99 THEN 420
30 INPUT "nmin";NMIN
40 INPUT "nmax";NMAX
50 XTITLE$="WAVELENGTH IN MICRONS"
60 INPUT "x title"; XTITLE$
70 SCREEN 2
80 LOCATE 24,35
90 PRINT XTITLE$;
91 LOCATE 1,2
92 PRINT NMAX
93 LOCATE 22,2
94 PRINT NMIN
95 LOCATE 10,1
96 PRINT "n"
100 LOCATE 1,20
110 PRINT YTITLE$
120 XLEN=600: YLEN=300
130 LINE(60,301)-STEP(XLEN,0)
140 LINE(60,301)-STEP(0,-YLEN)
150 XTIC=XLEN/11: YTIC=YLEN/10
160 DRAW "bm 60, 301"
170 FOR I=1 TO 11
180 LINE -STEP(XTIC,0)
190 DRAW "bu2 d4 bu2"
200 NEXT
210 DRAW "-bm 60, 301"
220 FOR I=1 TO 10
230 LINE -STEP(0,-YTIC)
240 DRAW "bi2 r4 bi2"
250 NEXT
260 LOCATE 23,5
270 FOR I=0 TO 11
280 XVAL=(4+I)/10
290 PRINT XVAL " ";
300 NEXT
310 LOCATE 1,1
320 FOR LMBDA=.4 TO 1.5 STEP .01
330 ON N GOSUB 1000,1040,1080,1120,1160,2000,2040
340 XVAL =60+(LMBDA-.4)*XLEN/1.1
350 YVALX=301-(NX-NMIN)*YLEN/(NMAX-NMIN)
360 YVALY=301-(NY-NMIN)*YLEN/(NMAX-NMIN)
370 YVALZ=301-(NZ-NMIN)*YLEN/(NMAX-NMIN)
380 PSET(XVAL,YVALX)
390 PSET(XVAL,YVALY)
400 PSET(XVAL,YVALZ)
410 NEXT
419 INPUT " ",AS: IF AS<>"END" AND AS<>"end" THEN 10
420 END

```

```

999 REM sucrose
1000 NX=SQR(1.8719+.446*LMBDA^2/(LMBDA^2-.0214)-.0113*LMBDA^2 )
1010 NY=SQR(1.9703+.4502*LMBDA^2/(LMBDA^2-.0238)-.0101*LMBDA^2 )
1020 NZ=SQR(2.0526+.3909*LMBDA^2/(LMBDA^2-.0252)-.0107*LMBDA^2 )
1030 RETURN
1035 REM niobate
1040 L=1/(LMBDA*LMBDA-.028)
1041 NX=2.21466264#+.0264414319#*L *(4.235971E-04)*L*L-.00600434126#*LMBDA*LMBDA
-(2.358354E-05)*LMBDA^4
1050 NY=NMIN
1060 NZ=2.14059953#+.0230733037#*L+(2.950437E-04)*L*L -.00507709036#*LMBDA*LMBDA
-1.23893E-05*LMBDA^4
1070 RETURN
1075 REM GfO
1080 NY=SQR(2.245*LMBDA^2/(LMBDA^2-.022693)+1)
1090 NX=SQR(2.24654*LMBDA^2/(LMBDA^2-.0226803)+1)
1100 NZ=SQR(2.41957*LMBDA^2/(LMBDA^2-.0245450)+1)
1110 RETURN
1115 REM TMO
1120 NY=SQR(2.27241*LMBDA^2/(LMBDA^2-.023359)+1)
1130 NX=SQR(2.273955*LMBDA^2/(LMBDA^2-.023333)+1)
1140 NZ=SQR(2.443016*LMBDA^2/(LMBDA^2-.025133)+1)
1150 RETURN
1155 REM calcite
1160 L=1/(LMBDA^2-.028)
1170 NX=1.64251430848#+.00574149845933#*L-.00005098207219#*L^2-.004052962915#*L
BDA^2+.00000573964755#*LMBDA^4
1175 NY=1.47853367021#+.00277070600522#*L-.00003344773321#*L^2-.0014417437971#*L
MBDA^2+.0001005603729#*LMBDA^4
1180 NZ=NMIN
1190 RETURN
1195 REM KTP
2000 NX=SQR(2.200009+.79993644#/(1-4.706234E-02/LMBDA^2)+1.320297/(1-100/LMBDA^2
))
2010 NY=SQR(2.300938#+.7225603/(1-5.793344E-02/LMBDA^2)+1.647279/(1-100/LMBDA^2)
)
2020 NZ=SQR(2.3469479#+.9690051/(1-5.793344E-02/LMBDA^2)+1.647279/(1-100/LMBDA^2
))
2030 RETURN
2040 REM sulfate
2050 NX=SQR(1.11209634#*LMBDA*LMBDA/(LMBDA*LMBDA-.0070301706#)+.107037023#*LMBDA
*LMBDA/(LMBDA*LMBDA-26.0656037#)+1)
2060 NY=SQR(1.1569400636#*LMBDA*LMBDA/(LMBDA*LMBDA-.0077005433#)+.11207305539#*L
MBDA*LMBDA/(LMBDA*LMBDA-12.0196206#)+1)
2070 NZ=SQR(1.104006217#*LMBDA*LMBDA/(LMBDA*LMBDA-.007909539122#)+.140500244#*L
MBDA*LMBDA/(LMBDA*LMBDA-14.90216041#)+1)
2080 RETURN

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