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OPTICS LAB -ECEN 5606

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Experiment No. 12

PHOTOREFRACTIVE CRYSTALS

1 Introduction

Photorefractive crystals have become the material of choice in recent years for recording dynamic volume holograms. They are highly sensitive nonlinear optical materials, with time constants between 1 millisecond and 100 seconds, making them appropriate for a wide range of applications. Diffraction efficiencies of 10-50% are readily obtainable in the ferroelectric oxides such as $BaTiO_3$, $Sr_xBa_{1-x}NbO_3$, and $LiNbO_3$:Fe, while lower diffraction efficiencies and higher speeds are characteristic of the paraelectric oxides such as $BiSi_{12}O_{20}$, BGO and BTO, and the semiinsulating semiconductors GaAs:Cr and InP:Fe.

2 Background

Photorefractive crystals are electrooptic photoconductive dielectrics which have sufficient populations of midband traps that can be optically excited and spatially redistributed. When light with energy below the band gap but sufficient to ionize the deep traps is incident on a photorefractive crystal, free carriers (usually assumed to be electrons) are produced that can redistribute due to influences of drift and diffusion, and become retrapped in unilluminated areas. This redistribution of the ionized traps produces internal space charge fields which locally modulate the index of refraction through the electrooptic effect, such that an incident optical wave can be influenced by the spatially modulated index of refraction.

The simplest experimental situation is when two coherent laser beams interfere in the volume of a photorefractive crystal to produce a sinusoidal interference profile in the $\hat{x}$ direction.

$$\vec{\mathbf{A}}_1(x,z,t) = a_1\hat{\mathbf{p}}_1 e^{i(k_x x + k_z z - \omega t)}$$

$$\vec{\mathbf{A}}_2(x,z,t) = a_2\hat{\mathbf{p}}_2 e^{i(-k_x x + k_z z - \omega t)}$$

Where a_i is the amplitude of the ith wave and $\hat{p}_i$ is the unit polarization vector of the ith wave. The interference profile is given by

$$I(x) = |a_1|^2 + |a_2|^2 + (\hat{p}_1 \cdot \hat{p}_2) 2a_1 a_2 \cos(2k_x x) = (|a_1|^2 + |a_2|^2)(1 + m\cos(2k_x x)) = I_0(1 + m\cos(K_G x))$$

where $m = (\hat{\mathbf{p}}_1 \cdot \hat{\mathbf{p}}_2) 2a_1 a_2 / (|a_1|^2 + |a_2|^2)$ is the fringe modulaton depth, and $I_0 = |a_1|^2 + |a_2|^2$ is the total intensity.

Free carriers are generated in proportion to the incident photon flux, and the available full traps which have an equilibrium density of $(N_d - N_d^+)$, where N_d is the total density of

donors and N_d^+ is the density of ionized donors [cm^{-3}]. The excited carriers recombine at a rate proportional to the free carrier density n [cm^{-3}] and proportional to the ionized centers locally available for recombination N_d^+, with a recombination rate constant γ_r [$\mathrm{cm}^{-3}/\mathrm{s}$],

$$\frac{\partial N_d^+}{\partial t} = (N_d - N_d^+)(sI + \beta) - \gamma_r N_d^+ n.$$

In this equation s [$\mathrm{cm}^{-2}/\mathrm{W}/\mathrm{s}$] is the photoionization crosssection and β [s^{-1}]is the thermal ionization rate, which is typically small compared to sI, and can be written as an effective dark intensity $I_d = \beta/s$. The time rate of change of the free carrier density is equal to the change of the ionized traps minus those carriers which leave a local volume element due to drift and diffusion:

$$\frac{\partial n}{\partial t} = \frac{\partial N_d^+}{\partial t} - \frac{1}{e}\nabla \cdot J,$$

where the current density J [A/cm^2] is given by the contributions of drift, diffusion and the bulk photovoltaic effect,

$$J_i = e\mu n E_i - k_b T \mu \frac{\partial n}{\partial x_i} + I\beta_{ijk}\hat{p}_j\hat{p}_k,$$

where β_{ijk} [A/W] is the bulk photovoltaic tensor. The internal space charge field is found from Poissons equation, once the trap densities and free carrier densities are known.

$$\nabla \cdot (\epsilon \cdot \underline{\mathbf{E}}^{\mathbf{sc}}) = \mathbf{e}(\mathbf{n} + \mathbf{N}_\mathbf{A}^- - \mathbf{N}_\mathbf{D}^+),$$

where $N_A^- = N_D^+(I = 0)$ is the compensating acceptor density which maintains charge neutrality. The modification of the dielectric impermeability due to the electrooptic effect is given by the tensorial expansion

$$\eta_{ij} = \frac{1}{\epsilon_{ij}} = \eta_{ij}^0 + r_{ijk}E_k^{sc},$$

where repeated summation notation is used and the electro-optic tensor $\underline{\underline{\mathbf{r}}}$ is tabulated in pm/V, which leads to a change of the index of refraction given by the standard relation,

$$\Delta n = -1/2n^3 \underline{\underline{\mathbf{r}}}\underline{\mathbf{E}}^{sc}.$$

The wave equation must be solved for the diffraction off this grating and the coupled mode solutions that result are usually of the form

$$\frac{\partial A_1}{\partial z} = -\frac{\alpha}{2}A_1 + \gamma\frac{A_1 A_2^* A_2}{I_0}$$

$$\frac{\partial A_2}{\partial z} = -\frac{\alpha}{2}A_2 + \gamma^*\frac{A_2 A_1^* A_1}{I_0}$$

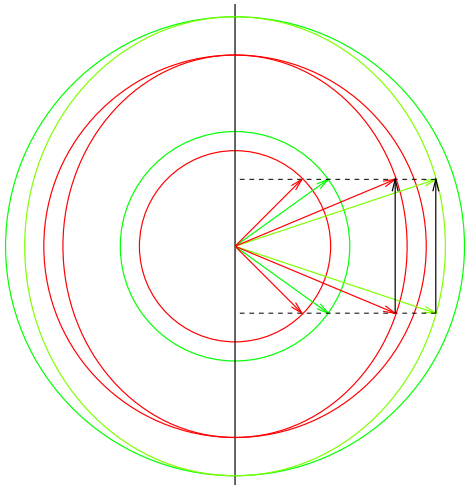
The complex coupling constant $\gamma = -i\pi\Delta n e^{-i\phi}/\lambda\cos(\theta/2)$ is characterized by a phase shift between the index grating and the interference profile given by ϕ. This phase shift is determined by the geometry and by material parameters. In $BaTiO_3$ and SBN, the diffusion terms dominate and the phase shift is close to $\pi/2$, which yields pronounced energy coupling between the beams; while in $LiNbO_3$ the photovoltaic fields dominate, producing a local response with $\phi \approx 0$ and little or no energy coupling.

3 Preparation

Read some of the photorefractive references in the lab.

Yariv, Optical Electronics, 3rd ed., Chap. 19.8
Hall et al, The Photorefractive Effect, Prog Quant. Elec., vol 10 (1985)
Gunter and Huignard, Photorefractive Materials, vol 1 and 2, Springer Verlag
Gunter and Eichler, Introduction to Photorefractive Materials

3.1 Prelab



1. When a volume grating is written with green light from an Argon laser at a wavelength of 5145 Angstroms and read out with red light from a HeNe laser at 6328 Angstroms, the angle of the red beam must be adjusted for optimal Bragg matching. When we use ± 10 degrees for the two Argon writing beams,what should the angle of incidence be for the HeNe beam in order to obtain optimal Bragg matching?

2. SBN:75 has a 4mm symmetry with reduced nonzero electrooptic coefficients of $r_{13} = r_{23} = 67\text{pm/V}$, $r_{33} = 1340\text{pm/V}$, and $r_{42} = r_{51} = 42\text{pm/V}$. For optical propagation in the xz plane, what direction of grating K_G vector should give the highest electrooptic coefficient? The photorefractive grating efficiency is actually decreased by large dielectic constants, as can be seen from Poissons equation, so that the grating diffraction efficiency is more nearly proportional to the direction dependent ratio r/ϵ. In SBN:75 $\epsilon_1 = \epsilon_2 = 500$ while $\epsilon_3 = 3400$, yielding a pronounced dielectric anisotropy. Find the optimum angle in the xz plane for the ratio r/ϵ. As a consequence, this is the angle at which the fanning becomes most prevalent.

3. Two wave mixing gain occurs in photorefractive materials because of the phase shift of the index grating with respect to the interferometric fringe profile. Show that the solution of the lossless intensity coupled mode equations (Eq. 1), are given by Eq. 2:

$$\begin{aligned} \frac{d}{dz}I_1 &= -\gamma_i \frac{I_1 I_2}{I_1 + I_2} \\ \frac{d}{dz}I_2 &= \gamma_i \frac{I_1 I_2}{I_1 + I_2} \end{aligned} \qquad (1) \qquad\qquad \begin{aligned} I_1(z) &= I_1(0)\frac{1 + r^{-1}}{1 + r^{-1}e^{\gamma_i z}} \\ I_2(z) &= I_2(0)\frac{1 + r}{1 + re^{-\gamma_i z}} \end{aligned} \qquad (2)$$

where $\gamma_i = \pi \Delta n/(\lambda \cos(\theta/2)) \sin\phi$ is the imaginary part (ie shifted by 90 degrees) of the complex coupling constant γ, and $r = I_1(0)/I_2(0)$ is the beam ratio at the input.

4 Procedure

This experiment can be done with $BaTiO_3$ or with SBN, and some parts can be done with $LiNbO_3$:Fe. Although the basic procedure is the same, the details will be different. Discuss which crystal to use with your TA.

4.1 Finding the c-axis

Either the 514nm Argon laser or 532nm double Nd:YAG laser can be used for this lab. Turn on the Argon laser, spatial filter and collimate the beam into a plane wave 3-5mm in diameter. Measure the power available, and under no circumstances apply more than a few W/cm^2 to the photorefractive crystal. A good working power is about 10mW in a mm^2 beam, but if you are focussing the beam with a lens it is easy to greatly exceed this power density, thereby permanently damaging the crystal. BE CAREFUL!!! Measure the polarization of the beam and determine if it is polarized parallel to the table (p = horizontal or extraordinary), or polarized vertically (s = ordinary). [Think about this, why is p extraordinary?] Place a half wave plate in the beam and rotate the waveplate to an angle that flips the polarization of the beam by 90°. With the waveplate in the path of the beam you will have one polarization, removing the waveplate gives you the other. Make the beam extraordinarily polarized. (Explain why this is the desired polarization). Aperture the beam down to $1mm^2$ with an iris, and if necessary, attenuate the laser to give no more than 10mW of power and direct this beam through the center of one of the crystal faces. Place a polarization analyzer on the output of the beam oriented to transmit ordinary polarization (ie crossed to the input). What do you see? Rotate the crystal by 90° through the four faces. When you see a distinct conoscopic zone plate pattern you have located the +c-axis as the direction of beam propagation. Remember this direction. Observe the crystal from the top as the incident beam propagates through the crystal a few degrees away from the +c-axis in both the vertical and horizontal planes (both tilted and rotated) Explain. (Look through a polarizer at this light scattered vertically.) You might want to see what happens when you illuminate the crystal with ordinarily polarized light. Remove the analyzer from the output.

4.2 Fanning

Rotate the crystal so that the c-axis is normal to the direction of propagation of the incident laser beam, which means that the c-axis and the polarization are collinear for extraordinarily polarized light. Wait 30 seconds; what do you see as time progresses? Rotate the crystal by 10 or 20 degees. Describe what you see. This process is called fanning and results from the amplification of randomly scattered light produced in the crystal through the phenomenom of 2 wave mixing. Rotating to an incident beam angle of about 45° should maximize this fanning effect in $BaTiO_3$. What happens to the time evolution of the fanning as you rotate to higher angles of incidence. What is the polarization state of the fanning? Change the incident beam to ordinary polarization; what happens to the fanning? Return to extraordinary polarization. Notice that the fanning pattern is unidirectional. Does it point towards or away from the +c-axis. Rotate the crystal to see.

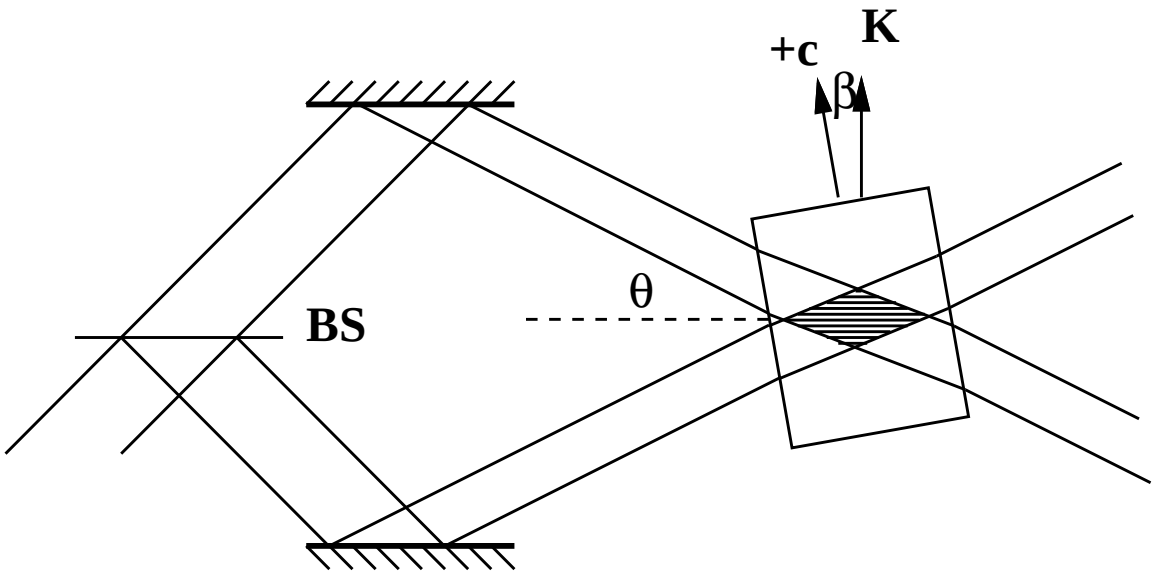
4.3 Writing a grating

Use a beam splitter to split the spatially filtered, extraordinarily polarized, Argon laser beam into two approximately equal beams, and use a pair of mirrors to form a symmetrical interferometer with between 10 and 20 degrees between the two beams. Place the intersection of the two beams on the crystal and adjust the beams so that they maximally overlap within the crystal. Make the crystal face normal to the bisector between the two beams, so that the grating vector is along the c-axis. Wait 30 seconds, block one of the beams, and look for a diffracted beam in the position that the blocked beam was previously occupying. In $BaTiO_3$ you should see a weak diffracted beam that slowly erases, while in SBN this geometry should give a strong diffraction. This is due to a holographic grating in the crystal that is diffracting the unblocked beam and reproducing the wavefront of the other. Rotate the crystal to a larger angle and record another grating by illuminating with both beams, making sure they still overlap in the crystal. Block one of the beams and watch the diffraction slowly erase. In $BaTiO_3$ it should be a much stronger diffraction in this orientation. Find an angle of rotation that gives a strong quick diffraction.

Measure the time constant of the erasure process by recording a grating, then blocking one of the writing beams while observing the diffraction with a detector, and noting the $1/e$ point in the exponentially decreasing diffraction. Decrease the incident internsity by a factor of 2 and measure the time constant. Decrease by 10 and 100 and measure the time constant. Plot the time constant for this erasure process as a function of the intensity of the erasure beams. How does it behave as a function of erase beam intensity. Using a variable beam splitter,equalize the power in the two writing beams and record a grating – be sure to wait long enough for the crystal to saturate (30 seconds). Measure the diffraction efficiency before much erasure has occured. Now make one of the writing beams 10,100, and 1000 times weaker than the other and write a grating, again waiting until saturation (60 seconds). Block the weak beam and notice that the diffracted beam is weaker than when the grating was recorded with equal intensity beams. This is because photorefractive crystals are sensitive to the contrast, or modulation depth, of the recording illumination which is defined as follows.

$$I(x) = I_0(1 + m\cos(\vec{\mathbf{K}} \cdot \vec{\mathbf{r}}),$$

where the total incident intensity is $I_0 = I_l + I_2$ and the modulation depth is $m = \frac{2\sqrt{I_1 I_2}}{I_0 + I_d}$, where I_d is a small thermally generated equivalent dark intensity. Neglecting I_d, for a beam ratio of 1:01, $m = .198$. The actual diffraction efficiency of the recorded grating is a nonlinear function of m, but to a good approximation it is nearly linear with m, especially in the regime $m < 0.8$.



4.4 Two wave mixing

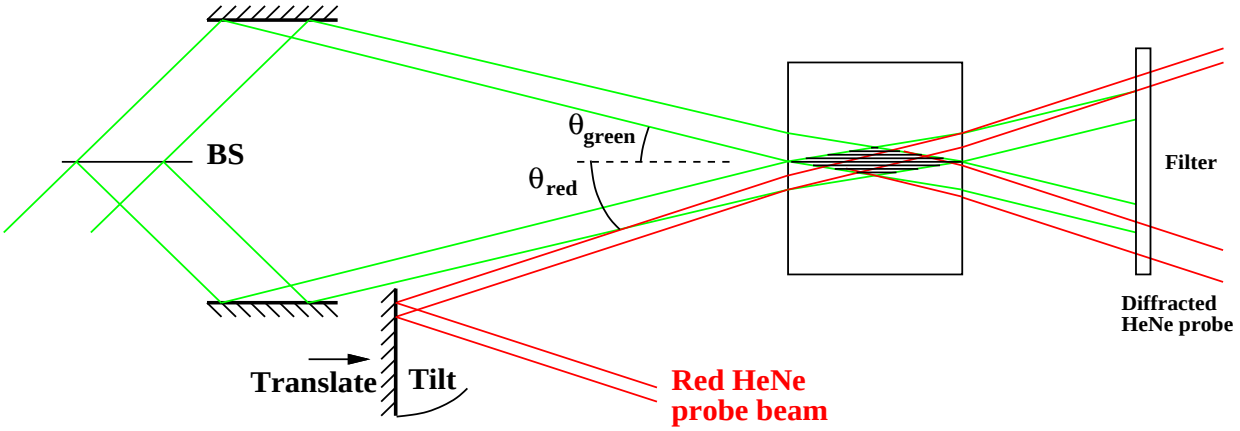
When two coherent beams illuminate the crystal simultaneously, a phase shifted index grating is dynamically formed by the charge transport mechanism driven by the spatial inhomogeneity present in the interference pattern. When we block beam 1 we can measure how much of beam 2 is diffracted into beam 1. However, since the gratings erase as we observe the diffraction, this is not the best way to measure the diffraction. While both beams are present on the crystal, beam 1 is being diffracted into beam 2; and simultaneously, beam 2 is being diffracted into beam 1. For photorefractive charge transport in the diffusion regime, the index grating is phase shifted with respect to the interference grating by 90°. When beam 1 is diffracted off the grating, it reproduces the wavefront of beam 2 exactly except for a 90° phase shift due to the phase shift of the grating, and an additional 90° phase shift due to the radiation from a polarization vector field, so the total phase shift is 180°. In this case the diffraction from beam 1 into beam 2 destructively interferes with beam 2, and the intensity of beam 2 decreases. At the same time beam 2 is diffracted into beam 1, and picks up a -90° phase shift due to the grating shift, which cancels the 90° phase shift due to the diffraction from a phase grating. This means that the diffracted component of beam 2 is in phase with beam 1, and constructively interferes so that the intensity of beam 1 increases. This effect can be observed by interfering two beams in the crystal and watching them exchange energy as the grating develops. A good geometry can be found by finding an angle that gives strong scattering of the pump beam, and then putting the signal beam in the region of highest amplification as mapped out by the fanning lobe. Begin with two beams of nearly equal intensity and notice which beam gets brighter and which gets dimmer. Observe the fanning lobe as you unblock the signal beam. Can you see any competition between fanning and 2 wave mixing amplification? Tap the table and notice that the beams blink on and off as the energy exchange goes back and forth due to the shifting of the interference fringe pattern with respect to the grating. Which beam picks up an energy gain – the one propagating slightly towards the +c-axis or the one propagating slightly away from the +c-axis? Now strongly attenuate the beam that gained intensity, and illuminate the crystal with only this weak signal beam. Measure the transmitted intensity in the absence of the pump beam. Now let the pump beam also illuminate the crystal, and measure the transmitted intensity of the

weak signal beam in the presence of the pump. The ratio between these two measurements is the two wave mixing gain. Repeat this measurement for several modulation depths, and plot the two wave mixing gain as a function of the signal beam intensity. You may want to try to get gain of the pump beam by attenuating it with respect to the signal beam. What happens in this case?

4.5 Image amplification

Place a transparency in the weak signal beam, with an intensity that gives a good two wave mixing gain. Image this transparency through the crystal onto an output observation plane using a lens system with the crystal near the Fourier plane (but not neccessarily in the Fourier plane because of the large peak powers there). Observe a magnified version of the output in the absence of the strong pumping beam. Then unblock the pump beam and observe the time development of the amplified image. Is the spatial frequency response of the image amplifier apparent?

4.6 Bragg matching a red probe beam

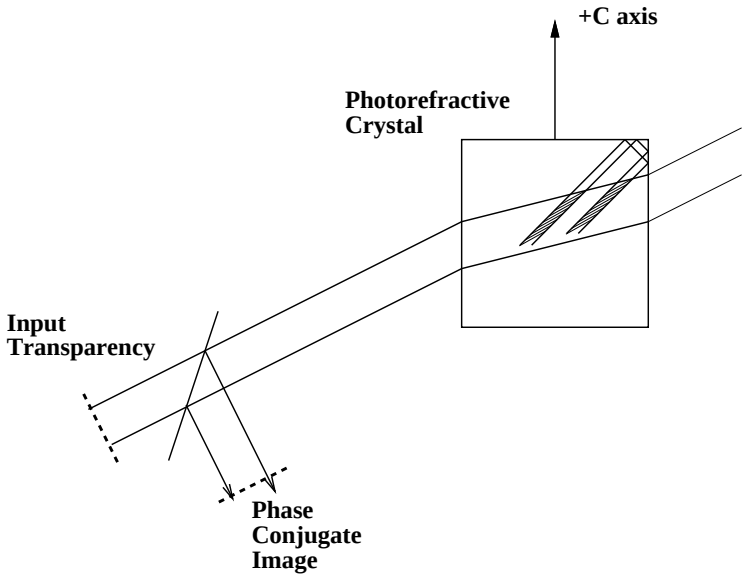


Using a mirror on a translation stage, align a red HeNe readout beam at an angle to read out the holographic grating written by the 2 green beams. Make sure that it is extraordinarily polarized, or it will not diffract. For a full angle of 20° between the two green beams this angle is $10° \frac{n_g}{\lambda_g} \frac{\lambda_r}{n_r}$, measured from the bisector of the two green beams. The Bragg sensitivity of this angle is only a few mrad (or a few arc minutes), so finding this alignment angle can be tricky. (Why is it so sensitive?) Notice that as you tilt the mirror to deflect the red beam to search for the correct angle the beam misses the crystal, so you have to iteratively tilt and translate the mirror appropriately. One approach is to write a grating with the green beams, then block the green beams while the weak red beam is illuminating the crystal, and rotate the crystal while looking for the diffracted red beam. Once you find the crystal rotation that gives the red diffraction, rotate the crystal back to the starting angle that is Bragg matched to the green beam, and use this information to figure out which way the red beam needs to be tilted and translated (i.e. higher angle or lower). Do this iteratively until you find the correct incidence angle that optimizes the diffraction of the red beam for the grating written by the green beams. Congratulations!

Now you can measure the time dynamics of the build up and erasure of the diffraction grating by placing your detector to intercept only the red diffracted beam (You may need a red filter as well). Erase the grating by blocking one of the green writing beams and/or

illuminating with the white light source. Now illuminate with both writing beams and watch the red diffraction build up. Block one of the beams and watch the red diffraction decay. You can now make a much more accurate measurement of diffraction efficiency vs. modulation depth, and you can measure grating recording time vs. incident intensity. Interestingly you can record a grating using ordinarily polarized green writing beams, as long as the red beam is extraordinarily polarized. This effectively eliminates dynamic beam coupling between the green writing beams which keeps the modulation depth constant throughout the crystal thickness, thereby simpifying the interpretation of the experimental results.

4.7 Self pumped phase conjugation



Place a beam splitter before the crystal in one of the beams at a 45° angle to the beam and place a card on the side of the beamsplitter that will be illuminated if the crystal produces a reflection. Illuminate the crystal with only that one beam at an incidence angle of about 20°, aligned so that the 1mm^2 diameter beam exits the back face about 1mm away from the $+c$ corner. Carefully watch the crystal from the top without touching the table, and observe the fanning filling the crystal with diffuse light. After about 10-30 seconds the fanning may collapse into a single beam that appears to go into the corner of the crystal from some point along the beam path through the crystal. This is the self-generated counterpropagating pump beam that reflects off the crystal corner back onto itself. This retroreflection reads out the grating which generated it and becomes a phase conjugated wave that will reflect back towards the laser, reflect off the beam splitter and illuminate the card. Place a transparency before the beam splitter and phase conjugate the image. Always wait 10-30 seconds after moving the crystal or any optic in order for the self pumped process to begin again. Experiment with this phase conjugation process and record your results. You may try some of the alternative self-pumped geometries if you wish.

Did you observe any spatial frequency limitations, and how did you measure them? What time response did you observe for the conjugation and how did this vary with incident illumination level?