"OPTICAL PHASE CONJUGATION IN PHOTOREFRACTIVE MATERIALS" (U)

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The photorefractive crystal barium titanate has been studied for its inherent physical properties and for its applications, including the locking together of separate laser beams. The output beams of two separate argon-ion lasers were locked together to within less than one Hz for an indefinite time. The technique uses four-wave mixing in the barium titanate crystal. A ring self-pumped phase conjugator was altered by inserting a nonreciprocal phase element in the ring so as to break its time-reversal symmetry. The device could be made to produce controlled frequency shifts and new output modes. Applications of this device are in mode conversion for optical computing. A transient detection microscope was invented which displays the images of moving objects, and which removes the stationary background. This all-optical device uses two-wave mixing in a barium titanate crystal.
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Since the demonstration of high-efficiency phase conjugation in barium titanate by Feinberg and Hellwarth in 1981, there has been increasing interest in the underlying physics of barium titanate crystals, and also in using phase conjugation to build new optical devices. An example of this recent interest in optical phase conjugation is the publication of separate articles, one written by a Soviet and the other by an American, in Scientific American magazine in 1985 and 1986. Additionally, the cover story of the March 31, 1988 issue of Nature magazine features a nonlinear optical microscope that uses photorefractive optics (and which was invented by the principal investigator and his students in the course of this research program). Also, an article about photorefractive nonlinear optics written by the principal investigator of this contract is the cover story of the October, 1988 issue of Physics Today magazine.

This final report describes the results of our research on the physics of photorefractive crystals, and the applications of these crystals to nonlinear optical devices. During this 3-year AFOSR contract we published 9 papers (in addition to conference proceedings) on important photorefractive materials questions and device applications. Some highlights are: We were able to identify and alter a key component (oxygen vacancies) in barium titanate, and then alter the photorefractive properties of the crystal by heat treatments. We were able to push the response time of barium titanate into the nanosecond regime, where we discovered that shallow traps play an important role in charge transport in this crystal. We were able to control the frequency shifts and alter the output modes of a self-pumped phase conjugator, and we produced a 16mm movie describing this project. We also invented a new kind of optical microscope that uses photorefractive optics: it preferentially displays the images of moving objects and automatically removes the images of stationary objects. Additionally, we made a videotape showing this microscope in operation.

We describe the goals and the results of this research project in more detail below.
RESEARCH OBJECTIVES
The original project goals were to:

1) Explore the use of optical phase conjugation for phase-locking the beams of independent lasers.

2) Alter the physical properties of photorefractive crystals by annealing them in an oxygen-rich or oxygen-lean atmosphere.

3) Explore the origin and the device applications of frequency shifts in self-pumped phase conjugators.

4) Explore the use of phase-conjugate interferometry to image fast events.

All of these goals have been met.

In addition to the above projects, we also were able to:

5) Demonstrate a device that converts one mode pattern into another using phase-conjugate optics.

6) Accurately measure the electrooptic and piezoelectric coefficients of barium titanate and strontium barium niobate crystals.

7) Determine the optical response of barium titanate to large optical intensities, and uncover the role of shallow traps in charge transport.

8) Complete two review articles on photorefractive materials and phase-conjugate optics: a long article published by Springer-Verlag and a short one published in Physics Today.

We describe our results in more detail in the following section.
I) RESULTS

1) Phase-locking separate lasers

We developed and demonstrated a new technique for phase-locking separate lasers using optical phase conjugation. This technique was highlighted in the July, 1986 issue of Lasers and Applications.

The principal reason for phase-locking lasers is to be able to efficiently combine the output of many small lasers into a single intense beam. For example, the peak intensity of an array of 1000 semiconductor diode lasers can be increased by a factor of 1000 if the lasers have been phase-locked.

Our technique for phase-locking lasers uses phase conjugation in a photorefractive crystal of BaTiO$_3$, and is shown in Fig 1b. We used two separate argon-ion lasers. The "master" laser beam was used to pump a BaTiO$_3$ crystal arranged as a self-pumped phase conjugator. The "slave laser" used the phase conjugator as one of its resonator mirrors, causing a beam to appear in the slave laser cavity. The output beam of the slave laser was automatically locked to the same frequency and phase as the beam from the master laser. The presence of the phase conjugator made the system insensitive to the precise position of any of the resonator mirrors. However, the system was sensitive to any vibrations faster than the time response of the phase conjugator (a few Hz in this case), so that the optical components had to be isolated from vibration. Further details and results are described in our paper published in Applied Physics Letters.

Recently Cronin-Golomb and Yariv demonstrated infrared phase conjugation in BaTiO$_3$ by cooling the crystal slightly below room temperature, suggesting that the technique described here can be extended to phase locking semiconductor diode lasers.

2) Altering the physical properties of photorefractive barium titanate

Barium titanate crystals have a large optical nonlinearity, but the response time of the crystal can be painfully slow when used with weak-intensity (milliwatt/cm$^2$) beams. However, the speed of the nonlinearity increases with intensity. Under a previous AFOSR project, we showed that the speed of barium titanate scales sublinearly with optical intensity, and we
FIG. 1. Locking lasers. (a) A master laser provides pumping beams to lock the optical oscillation of a slave laser by four-wave mixing in a nonlinear material. (b) A self-pumped phase conjugator increases the spatial overlap of the two pumping beams. (c) A self-pumped phase-conjugating mirror serves as the end mirror for both master and slave lasers. (d) Phase locking many lasers.
began work on altering the crystal's physical properties. In the present experiments we altered a barium titanate crystal using both reduction and oxidation treatments, and we were able to change the dominant carriers from holes to electrons, and to convert an inactive crystal into an active one, as shown in Figs. 2 and 3. Also, we discovered that the speed of the photorefractive effect scales faster with intensity when the crystal has been reduced (to make electrons the dominant charge carrier), as shown in Fig. 4. This has practical applications, because it implies that less intensity will be required to obtain high-speed response in these crystals.

We also proposed a physical model (shown in Fig. 5) to explain our results, and we showed the importance of oxygen vacancies in controlling the photorefractive properties of barium titanate crystals.

These results were published in the Journal of the Optical Society of America.3

3) Frequency shifts in optical phase conjugation.

Shortly after the principal investigator's invention of self-pumped phase conjugation using internal reflection in a barium titanate crystal,4 it was observed by the P.I. that the frequency of the phase-conjugate beam could differ by a few Hz from the frequency of the incident beam. In traditional four-wave mixing, where the pumping beams are externally supplied, the frequency of the signal beam is fixed by the frequencies of the three incident beams. However, in self-pumped phase conjugation, where all of the beams are derived from one incident beam, the frequency of the self-generated beams can differ from the frequency of the incident beam.

The P.I. previously demonstrated that these frequency shifts could be used to self-scan a tunable dye laser,5 and this self-scanning dye laser device was subsequently used for spectroscopy. However, the origin of the frequency shifts remained unknown.

In this project we performed experiments to demonstrate that, under some conditions, the efficiency of four-wave mixing in photorefractive materials can be increased by imparting a slight frequency shift to one of the incident optical beams. We first showed this theoretically, as seen in Fig. 6. This increase is the result of diminished competition between two-wave and four-wave mixing in the crystal. We then performed experiments using traditional four-wave mixing to demonstrate this enhancement, as shown in
Figure 2. The dominant charge carrier in a crystal of BaTiO₃ can be changed from holes to electrons and back to holes again. The function R measures the relative contribution of holes and electrons to the photorefractive effect. R can be altered from positive (holes dominate) to negative (electrons dominate) by heating the crystal in an oxygen-lean environment. Holes can be made the dominant charge carrier by heating the crystal in an oxygen-rich environment.

Figure 3. The effective density of photorefractive charge carriers $N_{pr}$ can be altered by treating the BaTiO₃ crystal at various oxygen pressures.
Figure 4. Plot of the photorefractive speed vs intensity for BaTiO$_3$ crystals that have been treated at different pressures of oxygen. Treatment at very low oxygen pressure causes the crystal speed to increase linearly with temperature.

Figure 5. Physical models of BaTiO$_3$ showing a hole-dominated crystal (top) and an electron-dominated crystal (bottom).
Figure 6. Plot of the calculated efficiency of four-wave mixing vs. the frequency shift $\delta f$ between the two writing beams in a photorefractive crystal. Note the increased efficiency at non-zero values of the frequency shift.

Figure 7. Measured efficiency for four-wave mixing vs. the frequency difference between the two writing beams in a photorefractive crystal of BaTiO$_3$. The efficiency shows a marked increase when the frequency difference is non-zero, as predicted by the theoretical curve above.
Fig. 7. These results suggest that a self-pumped phase conjugator may alter the frequency of its phase-conjugate beam in order to increase its four-wave mixing gain. (However, it is not clear that the beam intensities inside the barium titanate crystal are in the correct ratio to make use of this enhancement mechanism.) These results were published in Physical Review Letters.

4) Interferometry using real-time holography

We invented a microscope which removes the image of stationary objects from a scene, and shows only the moving objects. For example, if one views a section of living nerve tissue through this microscope, one should be able to see the slight changes in the nerve as an electrical signal propagates down the nerve. The microscope uses two-wave mixing between two coherent laser beams in a photorefractive crystal of BaTiO$_3$. One of the beams (the "image-bearing" beam) passes through the microscopic sample before it enters the crystal. The other beam (the "reference" beam), which is considerably more intense, is sent directly into the crystal, as shown in Figure 8. In the absence of any movement or change in the microscopic scene, the image-bearing beam and the reference beam interfere in the crystal and create a hologram. The beams then scatter off of this same hologram. The c-axis direction of the crystal is aligned so that the image-bearing beam is depleted by the interaction. The transmitted portion of the image-bearing beam is viewed with an eyepiece. In the absence of any change in the microscopic scene, only 1% of the original intensity of the image is transmitted through the crystal, so the image appears dark. However, if anything in the scene moves or changes, that portion of the image is immediately visible, as shown in Fig. 9. If the object stops moving, the hologram reforms, and the object fades from view, with the time constant determined by the intensity of the strong reference beam (typically about 0.2 second with an intensity of $I = 0.3$ W/cm$^2$). The contrast ratio of the microscope is about 100 to 1, and its resolution is limited only by the wavelength of light and the quality of the optical components, as in a conventional microscope.

Our article describing this "transient detection microscope" was published as the cover story of the March 31, 1988 issue of Nature. Subsequent news stories about this microscope then appeared in the New
Figure 8. The transient detection microscope. The incident laser beam is split into an object beam and a reference beam, which are made to intersect in the barium titanate crystal. The transmitted object beam contains the images of only the moving objects on the microscope slide.

Figure 9. Images of a resolution chart viewed through the transient detection microscope. The left image is dark because the chart is not moving. The right image was obtained immediately after the resolution chart was translated to the left. Note that both the old image and the new image are displayed simultaneously.
York Times and also in Science News. A patent disclosure has been filed with the University of Southern California.

We also made a 5-minute videotape showing the microscope in operation. This videotape was produced with the help of the USC video production department, and it shows protozoa swimming against a stationary background. When the optical processor is turned on, the background is removed, and any moving objects are clearly visible. (The sound track of the video has been re-mastered to improve its clarity, and a copy of this final videotape is included with this report.)

5) Dancing modes and frequency shifts in a phase conjugator.

In these experiments we deliberately altered a ring-type phase-conjugator in an attempt to control its frequency shift. In the process, we discovered that we could alter the output mode of the device, so that instead of "computing" the phase-conjugate replica of the incident beam, it computes a different output mode. This is the first demonstration of a stimulated process that computes an output beam that differs from a phase-conjugate beam. This ability to transform one optical pattern into another is essential to the future development of an all-optical computer.

The motivation for these experiments is to understand the origin of the frequency shifts observed in many kinds of "self-pumped" phase conjugators (i.e. phase conjugators that self-generate one or more of their pumping beams). Our approach was to modify the phase conjugator by controlling the direction or the phase of the pumping beams. Some phase conjugators are easier to modify than others. For example, the "CAT" phase conjugator is difficult to alter because all of its self-generated pumping beams are completely confined inside the photorefractive crystal by total internal reflection at the crystal faces. The ring phase conjugator is easier to alter because its pumping beams are directed into a ring outside of the crystal, where they are accessible. According to theory, the output of a ring phase conjugator should shift in frequency if the time-reversal symmetry of the ring is broken. We placed a Faraday rotator inside the ring and arranged quarter-wave plates on either side of the rotator so that the phase but not the polarization of the beams was altered, as shown in Fig. 10. The effect of these additional optical elements is to introduce a nonreciprocal phase change $\Delta \phi$. 
in the ring. By varying the magnitude of this phase asymmetry, we can control the frequency shift between the incident optical beam and the phase-conjugate output beam, as shown in Fig. 11. However, we found that when \( \Delta \phi \) was increased, the output mode becomes unstable, and a new mode appears and competes for the gain of the system, as shown in Fig. 12. These two modes are at slightly different frequencies, and so their interference pattern "dances" in real time. A further increase in \( \Delta \phi \) causes the new mode to win and the old phase-conjugate mode to disappear. Increasing \( \Delta \phi \) still further causes still higher-order modes to appear. When \( \Delta \phi \) approaches \( 2\pi \) the mode flips back to the phase-conjugate mode, and the cycle begins again.

These results were published in Optics Letters.\(^9\) In addition we made a short 16mm film called "Dancing Modes" about the experiment described above. The film uses animation, narration, and live footage filmed in the laboratory to describe and explain the action of the device. The film was made over a period of about thirty days with the help of seven graduate students from the Cinema Department here at USC, who served as production manager, director of photography, camera operator, editor, musicians, and animation cameraman. The film has been shown at numerous conferences, including a recent USA-USSR joint symposium. A copy of the film and a videotape (VHS format) were delivered to Dr. H. Schlossberg at the AFOSR.

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6) Measurement of the Pockels coefficients of barium titanate and strontium barium niobate.

We accurately determined selected Pockels and piezoelectric coefficients of the two most prominent photorefractive crystals, \( \text{BaTiO}_3 \) and \( \text{Sr}_{x}\text{Ba}_{1-x}\text{Nb}_2\text{O}_6 \). Our technique uses an interferometer shown in Figure 13. We also used a double-pass geometry in this interferometer to eliminate noise due to vibration of the crystal during the piezoelectric measurement. We were able to separately measure the piezoelectric and the Pockels coefficients, and we found that the Pockels coefficient of \( \text{BaTiO}_3 \) is 3 times larger than the values previously reported in the literature. (This discrepancy is due to previous workers' neglect of the piezoelectric expansion of the crystal during measurement.) Our new values for these constants indicate that there is marked competition between electrons and holes in almost all of the \( \text{BaTiO}_3 \) and SBN crystals studied to date. This implies that the optical nonlinearity of
Figure 10.
Optical setup to produce frequency shifts and dancing modes in a ring self-pumped phase conjugator.

Figure 11.
Frequency shift (lower trace) and output power (upper trace) of the output beam of the ring phase conjugator as a function of the nonreciprocal phase difference caused by the Faraday rotator. Note the abrupt changes in the frequency shift when the output beam hops into a new output mode.
Figure 12.
Output modes from the ring conjugator as the nonreciprocal phase shift is increased, from left to right. Increasing the phase shift to $2\pi$ returns the output mode to its initial phase-conjugate TEM$_{00}$ mode.
Figure 13.
Interferometer used to measure the piezoelectric (top inset) and Pockels (bottom inset) coefficients of photorefractive crystals.
these crystals could be greatly increased if this electron-hole competition were eliminated.

This paper was published in the IEEE Journal of Quantum Electronics.\textsuperscript{10}

7) High-speed phase conjugation with high-intensity beams in BaTiO\textsubscript{3}.

We are trying to determine the ultimate speed of the photorefractive effect in BaTiO\textsubscript{3} crystals, and we are continuing our measurements using intense pulses from an excimer-pumped dye laser. We first determined the optical damage threshold of the crystal to be typically 40 megawatts per cm\textsuperscript{2} at a wavelength of 578.7 nm. This is the intensity at which no surface damage was observed in any of our BaTiO\textsubscript{3} samples. Intensities ten times higher could also be used in some samples without damage. For a fixed intensity, the damage threshold increased as the spot size decreased.

While measuring the decay rate of photorefractive gratings in BaTiO\textsubscript{3} we found that this crystal contains additional levels and charge carriers that have been neglected in current theories of the photorefractive effect. These charge carriers produce anomalous behavior at high speeds. In particular, we found that a previously undetected shallow level near the conduction band, which, when populated, is capable of erasing, by thermal excitation into the band, any small-periodicity hologram that has been previously stored in the crystal. The density of these shallow traps varies markedly from sample to sample, which may account for the large difference (a factor of 10\textsuperscript{4}) in the dark conductivity of various samples of BaTiO\textsubscript{3}. Under intense illumination these shallow traps become saturated. Their presence accounts for the decrease in photorefractive sensitivity that we observed when using a high-repetition rate pulsed laser. The same shallow traps also cause "coasting" of light-induced gratings for hundreds of milliseconds after light beams are turned off in BaTiO\textsubscript{3} crystals.

Our paper describing these results has been accepted for publication in the December issue of Optics Letters.\textsuperscript{11}
8) Review articles on photorefractive materials and devices

An invited review article entitled "Photorefractive Nonlinear Optics" was written by the P.I. during this contract. This short article has been published as the cover story of the October, 1988 issue of Physics Today, a special issue devoted to lasers and their applications. The invited article describes the fundamental properties of photorefractive materials and devices, and contains photographs and descriptions of many of the projects supported by this contract (and also my previous contract with the AFOSR). AFOSR support is clearly acknowledged in this article.

In addition, the P.I. and his graduate student Kenneth MacDonald coauthored a book chapter entitled "PHASE-CONJUGATE MIRRORS AND RESONATORS WITH PHOTOREFRACTIVE MATERIALS" which has been published by Springer-Verlag in Chapter 5 of Photorefractive Materials and Applications, Volume 2. This long review article describes the fundamental physics and the applications of photorefractive materials, and the article contains acknowledgement of AFOSR support.
REFERENCES


II) PUBLICATIONS

Papers (and films) published under this contract:


3) Jack Feinberg and K.R. MacDonald, "Phase-conjugate mirrors and resonators with photorefractive materials," Chapter 5 of Photorefractive Materials and Applications, vol. 2, P. Gunter, Editor, to be published by Springer-Verlag, 1988. (This article is about 100 pages...)


Articles in the popular press about this work:

1) "Optical Phase Conjugation," in Scientific American, January, 1986 (see especially photographs of phase-conjugated cats and a spatula-laser).


4) Science News, April 16, 1988, "If it moves, catch it."

III) PROFESSIONAL PERSONNEL:

Principal Investigator:
Jack Feinberg

Post-doctoral Research Associates:
Robert M. Pierce
Daniel Bloch

Graduate Students:
G. David Bacher
Stephen Ducharme
Kenneth R. MacDonald
Daniel Mahger effteh
J.P. Jiang

Undergraduate students:
Aaron Seltzer

IV) ADVANCED DEGREES AWARDED:

Two students received Ph.D. degrees:
V) CONFERENCES


14. **Invited** to speak at the U.C. Irvine Undergraduate Physics Conference, "Reversing time with lasers," Irvine, California, April 5, 1986.


VI) INTERACTIONS WITH OTHER LABORATORIES:
University of Colorado:
   - Prof. Dana Z. Anderson - Photorefractive devices
   - Prof. Kristina Johnson - Phase-conjugating confocal microscope

Rockwell International Science Center:
   - Dr. Monte Khoshnevisan - Characterization of photorefractive materials
   - Dr. Ratnakar R. Neurgaonkar - Photorefractive crystal growth

New Mexico State University:
   - Prof. Marcus Cohen - Models of hologram competition in stimulated effects

VII) PATENTS OR INVENTIONS STEMMING FROM THIS RESEARCH EFFORT:
A patent disclosure on the "transient detection microscope" has been filed with the patent office of the University of Southern California.