

**TRANSIENT PHASE CONJUGATION**

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Final Report

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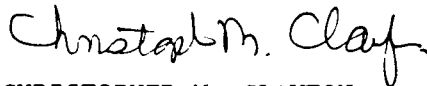
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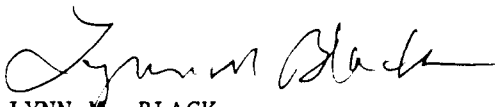
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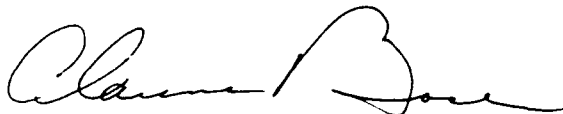


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13. ABSTRACT (Maximum 200 words) During the past 3 yr, material damage thresholds have been investigated as well as the magnitudes and response times of nonlinear susceptibilities of several important classes of optical materials. In addition, the Modified Twyman-Green Interferometer (MTGI) was invented. The MTGI determines both the magnitude and phase of the nonlinear susceptibility of a material relative to a known standard. This device has found significant acceptance within the optics research community. A phenomenon where energy may be exchanged between two frequency degenerate laser pulses has been characterized. Experimental investigations of optical phase conjugation via degenerate four wave mixing and beam coupling via degenerate two wave mixing using several nonlinear materials are compared. The components of the various physical phenomena which determine a nonlinear optical response as well as the interactions between these components are described. The strengths and weaknesses of the various optical materials and the utility of the MTGI are discussed. Finally, directions for future research are suggested.				
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INTRODUCTION

Optical Phase Conjugation (OPC) is a dimension of nonlinear optics which has been studied at an ever increasing rate for the past 20 years. Interest has been intensive due to the possible application of this and other closely related phenomena in the areas of optical telecommunications, optical computing (implemented digitally or as a "neural network"), image recognition and laser based weapons systems. In all of these potential applications, the OPC phenomenon is used as a source of optical feedback into a system. This feedback provides optical "error correction" and in some cases, optical amplification. Successful implementation has been achieved in several areas, including image recognition, image noise reduction and the improvement of laser beam quality during amplification stages.

In order for the OPC and related phenomena to be preferentially implemented over existing electronics technology, its advantages must be demonstrated. One advantage of photonic devices over electronic devices is that quanta of optical information travel comparatively independently and interact significantly only in carefully prepared, "nonlinear" devices.

The thrust of nonlinear optics in recent years has primarily been extensive materials science research to determine the dominant material parameters governing a nonlinear optical interaction, as well as to actually isolate materials which have a strong nonlinear reaction to irradiation without actually being damaged.

An important parameter which will, to a large extent, decide the extent of future optical implementation of optical and optoelectronic devices is the response speed, or "bandwidth" of an optical interaction or device. Ironically, the "speed of light" argument commonly used in favor of optical devices is not particularly significant since electronic signals propagate at close to light speed. In fact, for both optical and electronic devices, the response time parameters of a material is the primary governing parameter determining the effective bandwidth of a system. The bulk of



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recent experimental research suggests that there is an inherent trade-off between the "response speed" of an optical nonlinearity and its magnitude. At one extreme are materials such as the photorefractives; these crystals have very large nonlinear response magnitudes. For example, a coupling of two or more laser beams in a photorefractive results in a modulation (which is a function of the magnitude of the nonlinearity and the size of the interaction region) of one or more of the beams comparable to, or even exceeding, the intensity of the beams themselves for input beam intensities on the order of 1 mW/cm^2 in an interaction length of $\sim 1 \text{ cm}$. The bandwidth of the response is very low, with typical risetimes of 1 ms or more. At the other end of the spectrum are materials such as glasses and fiber optics. The materials typically have risetimes of only a few femtoseconds. A strong modulation of the input beams occurs only for input beam intensities on the order of 10^9 - 10^{10} W/cm^2 for interaction lengths of $\sim 1 \text{ cm}$. Fiber optics have circumvented the intensity requirement by easily extending the interaction lengths available in a laboratory setting.

Ideally however, the requisite interaction length should be in the range of 10^{-6} - 10^{-2} m to be technologically feasible and the corresponding average intensities should not exceed 1 W/cm^2 to minimize both power consumption and heat regulation problems. The damage threshold of a nonlinear medium or device is an additional important consideration in the search for materials.

During the past 3 years, material damage thresholds have been investigated as well as the magnitudes and response times of nonlinear susceptibilities of several important classes of optical materials. In addition, the Modified Twyman-Green Interferometer (MTGI) was invented, which is designed to characterize both the magnitude and the phase of a nonlinear susceptibility; this device has found significant acceptance within the optics research community. Furthermore, a phenomenon has been characterized by which energy may be exchanged between two frequency-degenerate laser pulses in a way which, until now, has received little or no attention.

In the following discussion of optical phase conjugation via degenerate four wave mixing (DFWM) and of beam coupling via degenerate two wave mixing, the results of experimental investigations on several materials are compared. The components of the various physical phenomena which determine a nonlinear optical response as well as the interactions of these parameters are described. The strengths and weaknesses of the various optical materials and the utility of the MTG interferometer are discussed. Finally, directions for future research are suggested.

DEFINITION OF THE PHENOMENON

A nonlinear optical response is defined as an intensity dependent modification of the characteristics, such as intensity or wavefront features, of an optical beam as it propagates in some medium. The influence of one photon on another takes place only indirectly and requires the presence of some matter in which intermediate states may be generated. In optically nonlinear materials, there are several physical mechanisms which may induce an optically nonlinear response. In many popularly used nonlinear media, two or more mechanisms may be simultaneously important. In categorizing a nonlinear response, one first determines whether it is the result of nonlinear absorption changes, such as saturation of absorption or "multiphoton absorption," or the result of intensity dependent phase changes such as self-focusing and self-phase-modulation. Either intensity dependent phase changes or absorption may be used to induce OPC. Both achieve this by creating a temporary diffraction grating in a material; this is generally known as a "transient grating."

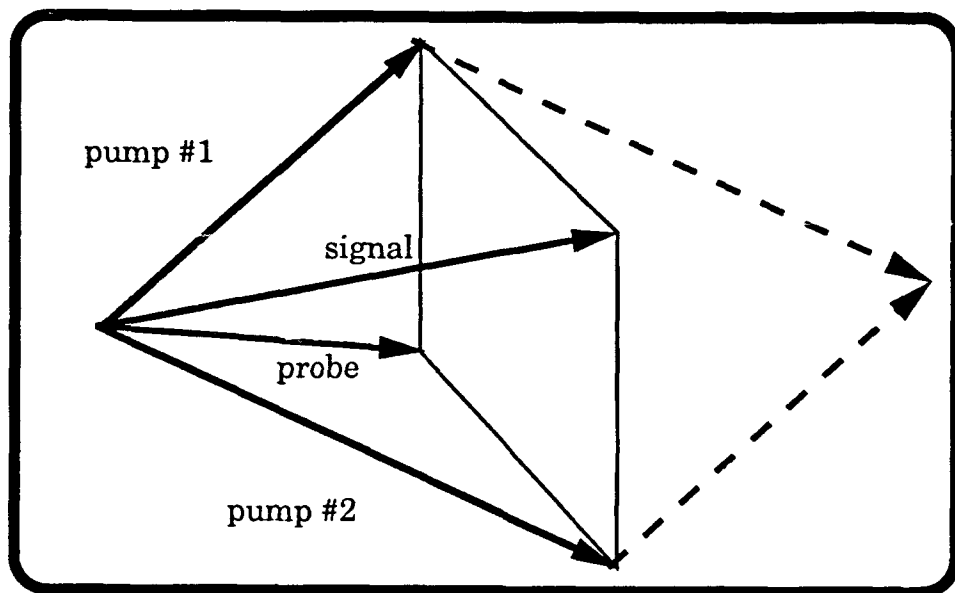
It is known from elementary optics that when a group of monochromatic "point" light sources is periodically arranged, the total energy of the light sources is channelled into specific directions; these directions are called the orders of diffraction. A diffraction grating is a physical device which has a periodic structure of some kind and, on illumination by an incident plane wave, introduces a periodicity into the phase of the reflected and

transmitted optical waves. The most commonly known "diffraction grating" is an array (on a piece of film or glass) of alternating light and dark slits that are separated from each other by a length on the order of a wavelength of the light to be diffracted. For optical frequencies, this length is on the order of 10^{-7} m. A diffraction grating can be made via nonlinear optics by overlapping two monochromatic laser beams in an appropriate sample. It is known that the overlap of two counter-propagating sinusoidal waves produces a standing wave, i.e., waves which have well-defined regions of spatially stationary high intensity and low intensity.

If two laser beams are crossed in a nonlinear medium, they will have some counterpropagating components which will form the regions of high and low intensity. Since the absorptive and refractive optical properties of a material depend on the intensity of light which passes through it, the material will have spatially periodic properties due to the presence of the intense standing waves formed by the laser beams. In a "saturable absorber" for example, the regions of high laser intensity may absorb only a finite quantity of optical energy in a given time. Optical energy in excess of this level will not be absorbed. The material therefore has alternating regions of high and low absorption; it becomes, for some finite time, a diffraction grating. Any additional incident optical energy will be "redirected" due to the presence of this grating.

In DFWM, two laser beams intersect in a nonlinear medium to form a "transient grating." Some fraction of a third beam is redirected by this grating. The fourth beam is defined to be the redirected component of the third beam. In phase conjugation by DFWM, this redirected beam travels precisely back along the direction from which one of the other beams was incident.

Figure 1 shows two different geometries for defining the phase-matched wavevector of the OPC signal in the case of frequency degeneracy in isotropic media. The loss of one photon from pump#1 and pump#2 is accounted for by an increase of one photon each for the probe and signal



$$\omega_{\text{signal}} = \omega_{\text{pump\#1}} + \omega_{\text{pump\#2}} - \omega_{\text{probe}}$$

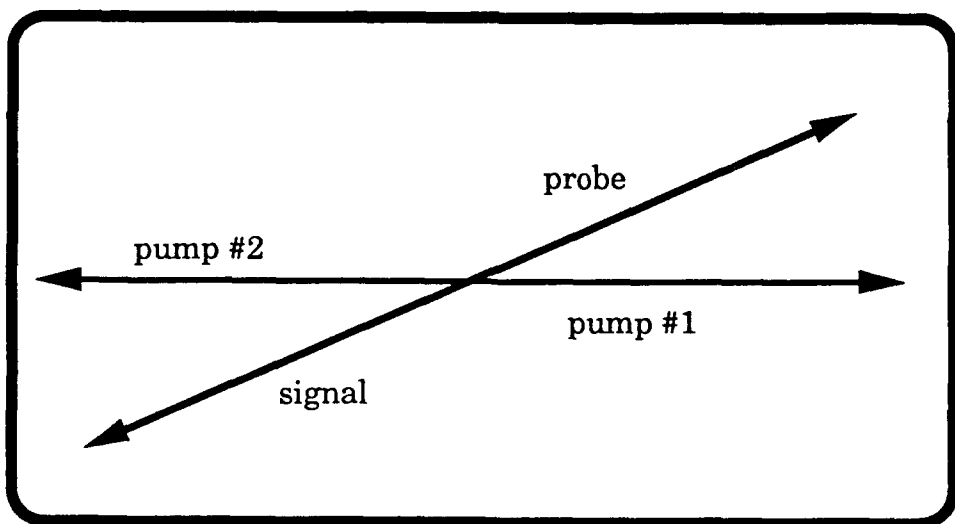


Figure 1. Energy and momentum conservation in four-wave mixing.

modes. The upper figure is generally known as the "boxcar" geometry while the lower figure produces OPC by DFWM. The boxcar geometry does not result in phase conjugation since the signal beam does not retrace the path of the probe beam. However, the geometry in the lower figure results in a signal antiparallel to the probe since the pump beams are antiparallel and

$$E_{\text{conj}} \propto \exp \left[i(\mathbf{k}_{\text{pump}\#1} + \mathbf{k}_{\text{pump}\#2} - \mathbf{k}_{\text{probe}}) \cdot \mathbf{r} \right]$$

For effective OPC efficiency, the two pump beams must be antiparallel to a high degree of accuracy. The intensities of the two pump beams must be equal since the effective wavevector in the nonlinear medium is intensity-dependent. If these conditions are provided and the frequencies of both pump beams and the probe are identical, an OPC signal will appear antiparallel to the probe beam for any intersection angle between the pumps and probe. It is preferable that the probe beam be almost parallel with one of the pumps in order to maximize the effective interaction length. In an applications environment, this would mean that the signal to be phase-conjugated should have a very small divergence so that OPC efficiency is uniform across the beam.

In phase conjugation by DFWM, this redirected beam travels precisely back along the direction from which one of the other beams was incident. A configuration in which the interaction of optical beams and a nonlinear medium results in an OPC signal is called a "phase conjugate mirror."

Figure 2 is a schematic representation of a comparison between a "normal" plane mirror and a "phase conjugate" mirror.

Thus, by retracing the path of a beam, the fourth beam will have precisely the optical characteristics that one of the other beams initially had; this is the basis of the noise removal and feedback qualities of phase conjugation which makes it potentially useful in optical technology.

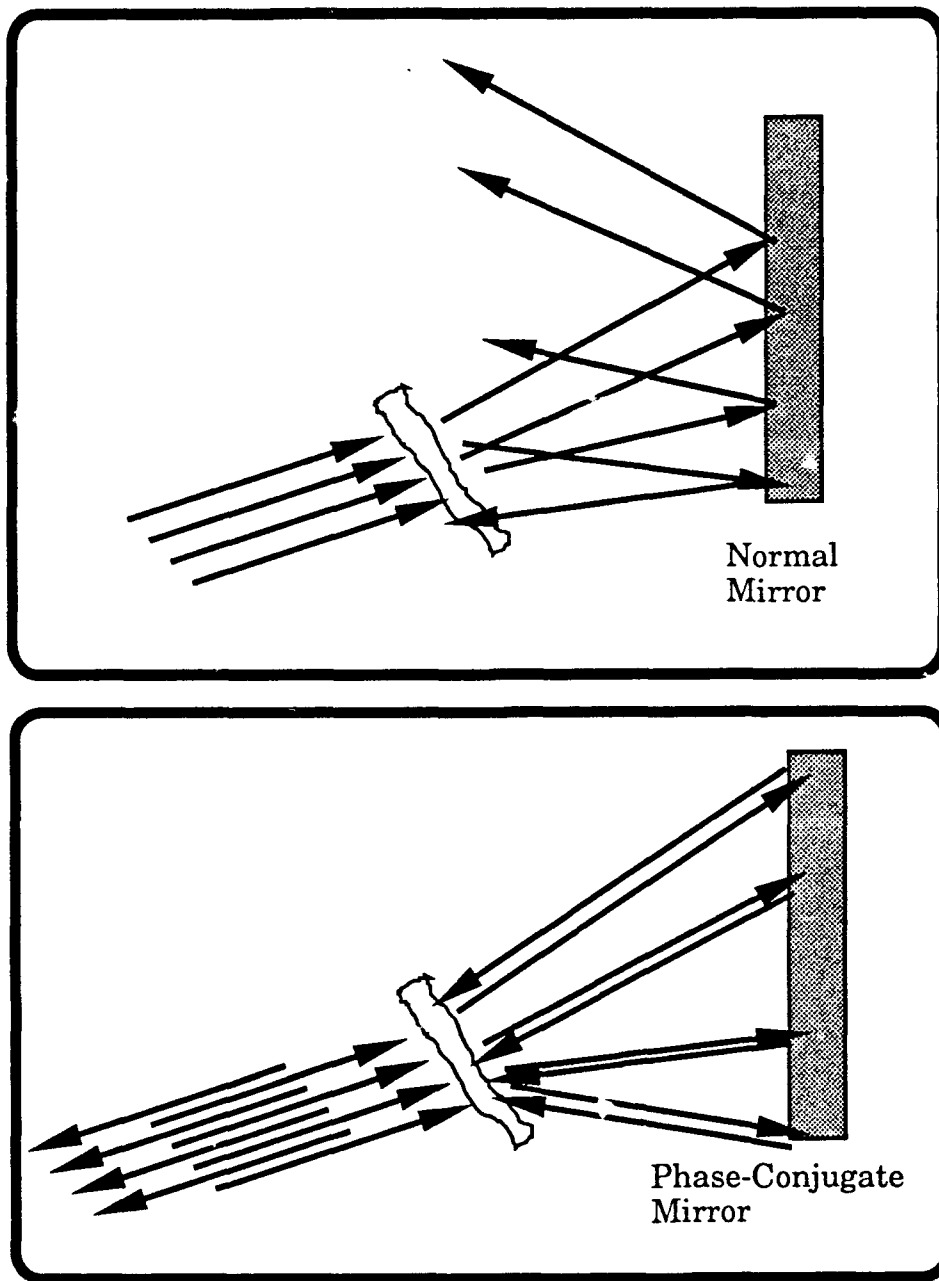


Figure 2. Phase conjugate mirror
vs. normal mirror.

It is important to characterize the quality (or fidelity) of the beam redirection, or phase conjugation, in different materials in order to designate materials which allow high efficiency phase conjugation (and sometimes amplification) of laser beams, especially under high duty cycle use. Ideally, a transient grating would diffract efficiently, would be created virtually instantaneously and would disappear immediately upon completion of its function. Since the characteristics of an optical beam change with time, the precise characteristics of the transient gratings should change with them as quickly as possible. It is also desirable that the creation of these temporary gratings do not cause any permanent change in the nonlinear material to increase the useful lifetime of the optical device.

PRELIMINARY INVESTIGATIONS

The first investigations of OPC by DFWM involved measuring the efficiency of high speed (picosecond) phase conjugation in carbon disulfide (CS_2) and a study of the influence of different experimental geometries for OPC on efficiency and response time measurements (Refs. 1 and 2). The picosecond experiments on CS_2 were designed to measure the influence of the intensity ratios between the three input beams in a DFWM experiment and to investigate the possibility of phase conjugation and amplification of laser pulses on a picosecond time scale.

Carbon Disulfide is a member of a group of optical materials called Kerr media. The observed nonlinear influence of laser beam intensity on Kerr-active substances is refractive in the visible spectral region. The gratings responsible for the OPC effect in these substances are transient phase gratings since the periodic refractive index of the grating induces a periodic phase change in the optical wavefronts as opposed to a periodic intensity variation. Kerr-active substances are among the fastest known optically nonlinear substances. These media often have subnanosecond response times and, in the case of CS_2 , the response time has a fast component on the order of femtoseconds and a comparatively slow component which responds in 1 ps.

The primary drawback of Kerr media as the basis of nonlinear devices is that comparatively high (10^9 W/cm²) peak laser intensities must be used to generate a significant efficiency of phase conjugation. A common measure of phase conjugate efficiency is called phase conjugate reflectivity and is defined as the ratio of output signal intensity to an input probe intensity.

In spite of the relatively low efficiency of the Kerr media, it was successfully demonstrated that a 25 ps, 2 mJ pulse could be phase conjugated and amplified if the appropriate ratios of input beam intensities were chosen; an OPC signal intensity of 3 times the input signal intensity was measured. The intensities of the two pump beams must be equal (to within <2 percent in these experiments) in order that the effect of "wavevector mismatch" is insignificant. Increasing the pump intensities did not improve the OPC efficiencies unless the pump intensities were very well matched. Also, the probe intensity should be <1 percent of the pump intensities in order that high efficiency and gain are achievable.

The investigations of the influence of wave mixing "geometry" on the OPC efficiency and response time measurements revealed that the use of a retroreflecting geometry (in which the second of the two pump beams is simply the retroreflection of the first after it passes through the sample) simplifies alignment, reduces complexity and provides a significantly more efficient generation of phase conjugate signals. However, for transient grating decay time measurements, results must be corrected to account for subtle changes in the beam overlap regions which occur when one laser pulse is delayed with respect to the others. These changes in beam overlap are a function of both the beam radius and the pulse duration when the laser pulse durations correspond to physical lengths comparable to or shorter than the sample dimensions (30 mm = 100 ps). These experiments were carried out on a CW modelocked Nd:YAG laser with a peak intensity of only 100 W/cm². At such low peak laser intensities, a material with a very high optical nonlinearity was required.

For these experiments, a Semiconductor-microcrystallite Doped Glass (SDG) was utilized as the nonlinear medium. This class of materials, a subset of a class known as microparticle suspensions, has comparatively high optical nonlinearities and response times ranging from milliseconds to picoseconds. These glasses have proven to be an important class of nonlinear materials and the physical phenomena at the origin of their optical nonlinearities have also proven to be extremely complex. To determine these properties, notable advances were made in the study of more general characteristics of optical nonlinearities.

One of the first steps in the determination of the nonlinearities in the SDG's was to measure the *phase* of the phase conjugate signal. As was discussed earlier, the induced transient gratings in nonlinear media may be periodic arrays of regions with varying refractive indices or varying absorption coefficients. The nonlinear refractive index of a medium is complex in nature as is the linear refractive index. Measurement of the phase of the phase conjugate signal determines the effective optical nonlinearity. Since the effective nonlinearity is often the result of a rapid succession of various physiochemical reactions, the phase measurement offers valuable insights into the various phenomena which are components of the optical nonlinearity.

An imaginary phase is associated with an absorption grating while a real phase is associated with a phase grating. Therefore, measurement of the phase of a signal shows the relative contributions of absorptive and refractive gratings to the total OPC signal and suggests the probable physical origins of the transient grating.

MTG INTERFEROMETER

A new type of interferometer, known as an MTGI (Fig. 3) was designed and developed to create a convenient system for the measurement of the phase of the OPC signals generated in various media. The sensitivity associated with this device has also made possible the measurements of

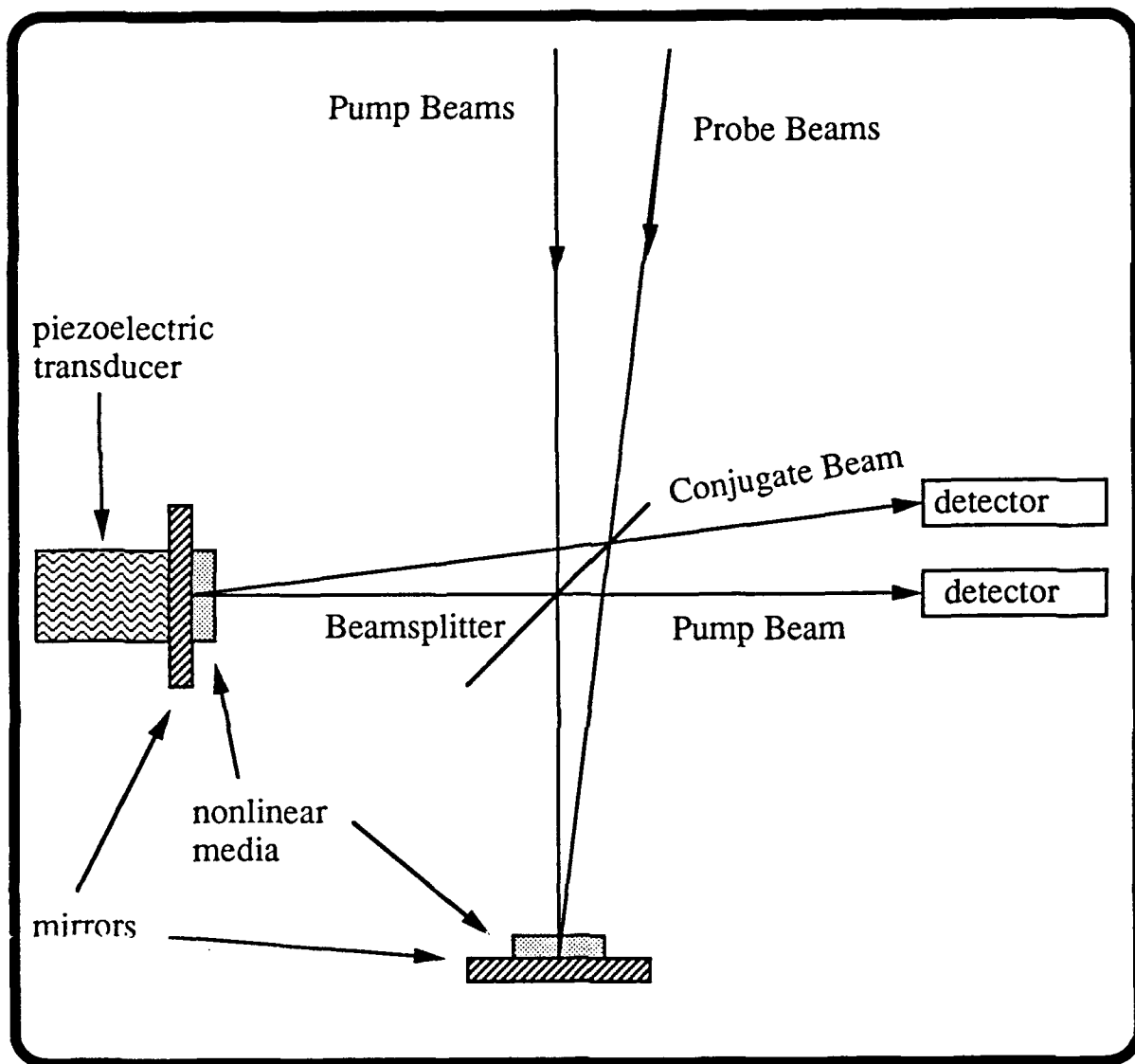


Figure 3. Modified Twyman-Green Interferometer with two phase conjugate reflectors.

OPC signals and nonlinearities which are ordinarily too weak to be detected by low power laser sources.

The MTGI has a standard two-arm interferometer geometry. On the retroreflecting mirror of each arm is a thin optically nonlinear sample. A "pump" laser beam is incident on a central beamsplitter; the resultant two beams pass through the sample, are retroreflected back through the sample and are recombined at the beamsplitter in standard interferometric fashion. Since the combinations of the pump beams, nonlinear media and retroreflecting mirrors form the bases of two OPC sources in the "retroreflection geometry," the addition of a probe beam fulfills the final requirement of a dual OPC system (each arm is an OPC source). Due to the geometry involved, if a probe beam intersects the sample at some small angle with the pump and is properly aligned to generate an OPC signal, then the alignment of the "second" of the two probes formed at the beamsplitter is automatic and simultaneous as is the generation of the second OPC signal. Since the two phase conjugate beams retrace the paths of the respective probe beams, they will recombine at the central beamsplitter and their interference as a function of optical path length difference may be measured by one detector while the pump beam interference is simultaneously measured by a second detector. It has been shown that the phase shift in the interference patterns generated by the two sets of beams is associated with the difference of the "absolute phase" of the nonlinear samples in each arm of the interferometer. If the absolute phase of one material is known, the phase of the second may be accurately determined.

Using this device, phase shifts of the SDG samples were measured; it was found that allegedly identical samples had slightly different absolute phases associated with them (Refs. 3, 4 and 5). Since the absolute phase is characteristic of a material, this discrepancy led to a comparison of several additional SDG samples and subsequently to a study of optically induced aging effects in the samples. This study considered the characteristic response times as a function of aging and the influence of

these effects on the intensity dependence of the phase conjugate reflectivity.

INFLUENCE OF LASER EXPOSURE ON OPC EFFICIENCY AND RESPONSE TIME

The aging of the SDG samples was a function of total exposure to laser radiation and of the rate of exposure to laser radiation (Ref. 6). While the overall phase conjugate reflectivity decreased irreversibly (at room temperature), the overall response times decreased as well. The OPC process resulting from low peak power, high average power lasers is strongly dependent on the imperfections and impurities (carrier "traps") of the semiconductor microcrystallites and therefore on the method of sample manufacture. The discrepancies in the laser intensity dependence of OPC efficiency, i.e., deviations from the "square law" intensity dependence could be explained (Refs. 7 - 9) by considering the physical origins of the optical nonlinearities and the relaxation times associated with these phenomena. In some regimes of interest, the coherence times of the pulses are much longer than the transient grating lifetimes in the samples. In this case, the integrated energy flux is responsible for the effective magnitude of the induced gratings, since gratings produced in successive short time intervals remain "in phase" with each other. This cumulative effect can cause deviations from the square law dependence in intensity if the grating lifetime is intensity dependent. Since it has been demonstrated that the induced grating is largely composed of a spatially periodic "filled trap" population and since it is also possible to saturate the traps, one can conclude that the reflectivity will deviate from a square law power dependence. In the opposite physical regime of interest, in which the coherence times of the pulses are much shorter than the grating lifetimes, the gratings induced in successive short time intervals "wash out" and the instantaneous value of the nonlinear optical susceptibility is probed. In this case, the intensity dependence of the OPC reflectivity will remain "square law" if the pump laser depletion and saturation of carrier excitation does not occur.

TEMPERATURE DEPENDENCE OF OPC IN SDG'S

The temperature dependence of the OPC efficiency was discussed (Ref. 10). This study showed that, since the lifetimes of the trapped carriers responsible for the variation in the refractive index depends on the temperature, low temperatures increase the transient grating lifetime and thus the equilibrium transient grating magnitude. The OPC process in SDGs can be made significantly more efficient, i.e., less input power required for a given OPC output intensity magnitude when the temperature is lowered. However, the response times of the transient gratings increased, thus lowering the effective "bandwidth."

METALLIC SUSPENSIONS

The microparticle suspension investigations were not limited to the SDGs. Metallic gold colloidal suspensions in liquid form and in solid form were also studied (Ref. 11). The metallic colloids are considered to be attractive candidates as optically nonlinear media because of their enormous third order nonlinear optical susceptibilities. The experiment utilized suspensions containing a volume fraction of 4×10^{-6} of the metallic particles, with the remaining volume of the sample being primarily water. The experiments showed that, for gigawatt per square centimeter input pump intensities, the OPC efficiency for the gold suspensions was 10^{-3} of that of CS_2 , the standard of high speed nonlinear media. After correcting for the small volume fraction of the gold, it was found that the $\chi^{(3)}$ of the gold was effectively $\sim 10^4$ times that of the CS_2 . The linear absorption of the metallic particles is also extremely strong. The magnitude of the linear absorption precluded any increase in volume fraction and that the laser intensities required to produce even relatively small OPC efficiencies (reflectivities of 10^{-4} - 10^{-3}) was sufficient to cause permanent damage to the samples. As was the case with the SDG samples, the damage depended on both the instantaneous laser intensities and on the total accumulated energy flux.

OTHER COMPOSITE SYSTEMS

The most recent OPC investigations[†] have been studies of azo dye-molecule-doped liquid and solid polymer matrices. These systems show significant promise as potentially useful nonlinear media. They have proven to be exceptionally easy and inexpensive to prepare and they provide a significant nonlinear response even with relatively low laser input intensities ($\sim 10 \text{ mW/cm}^2$ average intensity). Additionally, the response times typically associated with them are less than millisecond scale and sometimes less than microsecond-scale. Although the effective nonlinearity originates from the dye molecules (no OPC signals were detected in samples of pure, undoped matrices), the interaction of the excited dye molecules with the matrix provides a significant component of the effective transient grating. The OPC signal characteristics depend strongly on the solvent matrices; there were qualitative differences in the OPC signal characteristics originating from the liquid and the solid matrices although they were chemically identical otherwise. This is further evidence that the effective optical nonlinearity is a rather complex function of several simultaneous physical processes. Preliminary investigations of these systems revealed that the quantum chemistry involved is also complex and not completely understood. The intensity dependence and temperature dependence of the OPC signals from these materials suggest the influence of conformational changes in the dye molecules and the influence of triplet state kinetics connecting the conformations. It is evident that there remains a significant amount of work to be done to completely characterize the physics and chemistry of

[†] Tomov, I.V., Dutton, T.E., VanWanterghem, B. and Rentzepis, P.M., "Optical Phase Conjugation in Polymer Systems," accepted by the SPIE.

Tomov, I.V., VanWanterghem, B., Dvornikov, A.S., Dutton, T.E. and Rentzepis, P.M., "Degenerate Four Wave Mixing in Azo Dye Doped Polymer Films", accepted by J. Opt. Soc. Am. B

Tomov, I.V., Dutton, T.E., VanWanterghem, B. and Rentzepis, P.M., "Temperature Dependence of Degenerate Four Wave Mixing in Azo Dye Doped Polymer Films," accepted by J. Appl. Phys.

these substances before the engineering of an optimum configuration may be achieved.

BEAM COMBINATION BY DEGENERATE TWO WAVE MIXING

The nonlinear optics projects have not been exclusively OPC by DFWM. Experimental investigations of degenerate two-wave mixing in isotropic media have recently been carried out (Ref. 12).

The coupling of two frequency-degenerate beams has been the subject of extensive research for several years. Until very recently however, beam coupling was accomplished only in anisotropic crystals, typically photorefractives.

As was mentioned early in this report, photorefractives have frequently been used as the active element in nonlinear optical experimental systems. The optical anisotropy of the crystal allows one to break the symmetry of two degenerate beams; by adjusting the orientation of the crystal relative to the two beams, a preferred direction of energy transfer can be assigned and the direction of energy flow between two otherwise identical laser beams can therefore be controlled.

Two beam coupling via the photorefractives is subject to the same limitations placed on all photorefractive-based nonlinear optical devices, namely, relatively slow response times. Typically, response times are on the order of 10^{-4} - 10^0 s. Although some photorefractives have significantly faster response times, the direction of energy transfer is still a function of crystal orientation.

Two-beam energy transfer on a picosecond time scale has been achieved.* Using CS_2 as the active medium, energy was successfully

* Dutton, T.E., Rentzepis, P.M., Scholl, J., Shen, T.P. and Rogovin, D. "Picosecond Degenerate Two Wave Mixing in Isotropic Media" submitted to Jour. Appl. Phys.

transferred between two identical laser pulses. The maximum measured gain has been on the order of 10 percent for identical laser pulses of gigawatt per square centimeter intensity. By increasing the ratio of input intensities of the two beams, this gain can be enhanced.

The direction and magnitude of energy transfer in this technique depends on the polarizations of the two laser beams and on the elapsed time between their passages through the active medium (which should have a relaxation time comparable to the laser pulse durations). The required variability in the delay range is on the order of the length of the pulse. The experiments involved pulses with a 25 ps duration; this corresponds to a required optical path length variability range of ~ 10 mm to control the direction of the energy transfer. An extension of the technique to femtosecond pulses would lower the required path variability proportionally.

CONCLUSIONS

Significant advances have been made in the understanding of the interplay between the magnitude of a nonlinear optical response and the characteristic relaxation times associated with them. Advances have also been made in the techniques used to study these phenomena and in the characterization of several classes of nonlinear optical materials.

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