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DIVISION OF ENGINEERING AND APPLIED PHYSICS HARVARD UNIVERSITY - CAMBRIDGE, MASSACHUSETTS Office of Naval Research Contract Nonr-1866(49) NR - 015 - 807 ARPA Order No. 455

#### SEMIANNUAL TECHNICAL SUMMARY REPORT

Covering Period December 1, 1963 - June 1, 1964

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#### SEMIANNUAL TECHNICAL SUMMARY REPORT

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#### SEMI-ANNUAL TECHNICAL SUMMARY REPORT

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#### Synopsis

The nonlinear optical constants of several III-V semiconductor compounds have been determined at  $\lambda = 1.06$  microns with a neodymium glass laser, to supplement the data taken with the ruby laser. Dispersive properties of the nonlinear susceptibility have been established.

The coupling between vibrations and light waves in Raman laser media has been studied theoretically. Some new results have been reported in the May 4, 1964 issue of the <u>Physical Review Letters</u>.

The experimental set-up has been built to send a laser beam and a Stokes beam of separately controlled intensity and direction through a thin sample cell. In this manner, accurate measurements of the Stokes gain as a function of angle, polarization and intensity of the laser beam will be made possible. The same method can be extended to the measurement of anti-Stokes gain.

#### A. Nonlinear optical constants of III-V compounds

The measurement of the second harmonic production in reflection from GaAs, GaSb, InAs and InSb single crystals has been extended to a longer wavelength by the use of a neodymium glass laser. The nonlinear susceptibility is found to be twenty to fifty percent smaller at  $\lambda = 1.06$  microns then at  $\lambda = 0.694$  microns. This result can be understood on the basis of theoretical expressions for the nonlinearity in the presence of band absorption

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at the fundamental and/or second harmonic frequency. The case of GaAs is especially interesting, because the laser at  $\lambda = 1.06\mu$  is not absorbed. In this case, the nonlinearity can also be determined from second harmonic production in transmission. This result agrees with the reflection measurement. The transmission method is, however, less accurate, because it depends more sensitively on the difference in the linear dielectric constants at the fundamental and second harmonic frequency. The nonlinear susceptibility does <u>not</u> change drastically as the fundamental frequency is varied from just above to just below the absorption edge. An order of magnitude change in the nonlinearity is expected to occur when the second harmonic frequency is varied across the gap.

This material has been presented at the March 1964 meeting of the American Physical Society in Philadelphia, Pennsylvania, and has been accepted for presentation at the International Semiconductor Conference, to be held in Paris, July 20 - 24, 1964. An invited paper was given before the Optical Society of America, April 2, 1964 in Washington, D.C.

#### B. Theory of nonlinear Raman-type susceptibilities

The coupling between laser, Stokes and anti-Stokes light waves and the molecular vibrations in Raman laser media has been investigated theoretically. A careful analysis of the coupled wave equations shows that neither the Stokes nor the anti-Stokes wave has an exponential gain factor in the direction of exact momentum matching. On either side, just off this direction, exponential gain for one wave with a mixed Stokes-anti-Stokes

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character occurs due to parametric coupling with the laser beam. These interesting conclusions have been reported [1] and further details may be found in this reference.

The theory is being extended to the case of higher order Stokes and anti-Stokes frequencies. Furthermore, it has been found, experimentally as well as theoretically, that depletion of the laser beam plays an important role. The laser field must not be treated as a fixed parameter and the wave at the laser frequency should be included explicitly in the set of coupled amplitude equations. Numerical work on this nonlinear problem is now in progress.

#### C. Experimental measurement of Stokes and anti-Stokes gains

The theoretical considerations have helped to design an experimental arrangement for reliable measurements of Stokes and anti-Stokes gains. A Q-switched ruby laser beam is split into two beams of about equal intensity by a beam splitter. One beam creates Stokes light in a Raman cell. With the aid of two selective dielectric mirrors this Stokes beam is superimposed on the other laser beam. By rotation of one of the mirrors the angle between the laser and the Stokes beam can be varied. The intensity and polarization of either beam can be adjusted independently by suitable absorbers. The superimposed beams pass through a second Raman cell, the length of which is chosen so that the gain in this cell is about a factor of three or less. Preliminary tests have confirmed the usefulness of this scheme. Reliable measurements of the Stokes and anti-Stokes gains in a number of liquids will be carried out in the next period.

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#### D. The inverse Faraday effect

When an intense circular polarized light beam passes through a transparent medium, a dc magnetization is produced along the direction of propagation. This is the magnetic analogue of the dc rectification of light, reported by Franken [2]. As this latter effect may be regarded as the thermodynamic inverse of the linear electro-optic Kerr effect, so can the dc magnetization be considered and calculated as the inverse of the Faraday effect [3].

A circular polarized Q-switched ruby laser beam falls on a  $CaF_2$ crystal doped with rare-earth ions, e.g.,  $Eu^{2+}$ . A pick-up coil followed by a wide band amplifier detects the magnetization pulse produced by the light pulse. Preliminary experiments have indeed shown the presence of a signal, the sign of which depends on the sense of circular polarization of the laser beam. The dependence of this signal on concentration and nature of the paramagnetic ion, on temperature, and on a dc magnetic bias field is at present under investigation.

#### References

- 1. N. Bloembergen and Y. R. Shen, Phys. Rev. Letters 12, 504 (1964).
- 2. M. Bass, P. A. Franken, J. F. Ward, and G. Weinreich, <u>Phys. Rev.</u> Letters 9, 446 (1962).
- 3. P. S. Pershan, Phys. Rev. 130, 919 (1963).

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#### Nonr-1866(49)

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