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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A variety of nonlinear optical processes in semiconductors, have been investigated experimentally and theoretically. Among them are: nonlinear optical-impurity interactions, spin-induced four-photon mixing, FIR difference frequency generation, parametric excitation of plasma waves, nonequilibrium carrier dynamics under intense laser excitation, and enhanced four-photon mixing in the Gunn effect regime. The ultimate goals of this work are the development of tunable IR sources; and the understanding of carrier dynamics in semiconductors.

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*Infrared Nonlinear Optics in Semiconductors*I. Research Objectives*AFOSR-76-2894*

The purpose of this program has been to investigate a variety of infrared, nonlinear optical processes in semiconductors. The investigations have combined theory and experiment. Generally speaking, the processes studied have involved mobile or weakly bound electrons in these materials. Among them are:

- A. Nonlinear optical-impurity interactions in Ge and Si.
- B. Nonequilibrium carrier dynamics in n-Ge under intense laser illumination.
- C. Resonant, spin-induced, four-photon mixing in (Cd,Hg)Te.
- D. Resonant, spin-induced FIR difference frequency generation in InSb and (Cd,Hg)Te.
- E. Optical properties of n-InSb samples with layered doping.
- F. Stimulated excitation of collective modes in n-InSb, n-(Cd,Hg)Te and other narrow gap materials.
- G. Studies via four-photon mixing of carrier dynamics in the Gunn effect regime in GaAs.

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II. Status of Research Effort

Facilities

During the first year of this grant a laboratory containing equipment for performing infrared nonlinear optical experiments in semiconductors was established. This facility now includes:

- i. two simultaneously Q-switched CO₂ lasers with peak powers of 3 KW,
- ii. spectrometers,
- iii. IR detectors,
- iv. magnets, including a 70 Kg superconducting system,
- v. fast electronics,
- vi. optical tables, miscellaneous optical components, etc.

The total value of the equipment is about \$120K, of which \$60K was provided by AFOSR, \$21K by MIT. The rest was borrowed, scrounged, etc.

The laboratory has been very successful. It is now used almost daily, by students, staff, and faculty, to perform IR nonlinear optical experiments in semiconductors.

Research Projects

A. Nonlinear Optical-Impurity Interactions in Ge and Si

The first successful experiment in the IR laboratory was the observation of resonant, impurity-induced, four-photon mixing in n-Ge. The Ge crystal was pumped with two CO₂ laser beams at frequencies ω_1 and ω_2 . Power at frequency $\omega_3 = 2\omega_1 - \omega_2$ was monitored as a

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function of the difference frequency, $\Delta\omega = \omega_1 - \omega_2$. Sharp resonances were observed when $\hbar\Delta\omega$ equalled the valley-orbit splitting of the donors. This effect has been observed in Ge:P, Ge:As, and Si:P. Until recently, tuning was achieved by using different pairs of CO₂ laser lines - a clumsy, inaccurate method. Lately, it has been demonstrated that the resonances can be observed with magnetic field tuning of the impurity energy levels. This technique will permit accurate measurements of energy level position vs. field. Theoretical calculations of the energy level structure are nearing completion; detailed experiments are in progress. This work will provide information concerning:

- i. diamagnetic compression of donor wave functions,
- ii. level anti-crossings due to off diagonal matrix elements of the Zeeman interaction,
- iii. donor-donor interactions,
- iv. excitation transport in donor systems.

The theory of impurity-induced four-photon mixing has been worked out in detail, and is in good agreement with experiment. These studies determine impurity Raman cross-sections.

B. Nonequilibrium Carrier Dynamics

The four-photon mixing experiments (described above) imply large impurity Raman cross-sections. This result led, in turn, to an attempt to demonstrate stimulated, impurity Raman scattering in n-Ge. The experiment failed because the intense laser beams required stripped electrons from the donors. The carrier excitation process

has since been studied via measurements of optical absorption vs. laser power. In cold n-Ge crystals, the optical absorption decreases markedly (by more than a factor 10) in the power range $1 - 10 \text{ MW/cm}^2$. A kinetic model, which balances the photoionization rate from IS levels against the rate of carrier recombination with donors, gives an excellent fit to the data. These measurements determine the photoionization cross section, the donor recombination cross section, and the thermalization rate of hot carriers in n-Ge.

C. Resonant, Spin-Induced, Four Photon Mixing

Extensive studies of resonant, spin-induced, four photon mixing have been performed in (Cd,Hg)Te samples provided by Honeywell. In general, one observes several distinct spin resonances - a result indicating that the crystals are inhomogeneous. Such experiments are now being used by the Honeywell group as an analytic tool to determine g-valley and alloy composition as a function of position in (Cd,Hg)Te.

The MIT work has focussed on homogeneous portions of the (Cd,Hg)Te crystals, which show exceedingly sharp ($Q \sim 500$) spin resonances. At higher laser powers (in the 500 W/cm^2 range) the resonances broaden and saturate. The values of the spin relaxation times, T_1 and T_2 , inferred from these measurements are comparable to those found in n-InSb.

A student supported by this grant, Asif Khan, spent the summer of 1978 at the Honeywell Laboratory in Minneapolis making further studies of IR nonlinearities in (Cd,Hg)Te. Among the new processes

he observed were spin-induced six-photon mixing and polarization-forbidden four-photon mixing. Khan has since become a staff member at Honeywell. He interacts regularly with the MIT IR group and also furnishes (Cd,Hg)Te samples.

D. Resonant, Spin-Induced FIR Generation

The aim of this project is to generate tunable FIR via difference frequency generation. n-InSb samples, with spatially modulated doping, are the ideal mixing medium for such experiments. Until such crystals become available, it is planned to test the feasibility of the technique with difference frequency generation in bulk (Cd,Hg)Te. This material is preferable to InSb because its bandgap can be matched to CO₂ lasers, whereas 5 μ pumping is required in InSb. High field linewidth and absorption measurements are now being performed in (Cd,Hg)Te samples to determine the optimal configuration for the FIR experiment.

E. Optical Properties of n-InSb Samples with Modulated Doping

Optical reflectivity has been used to characterize n-InSb samples, with modulated doping, grown by Prof. Witt's group. These crystals are far too heavily doped ($n \approx 10^{18}/\text{cc}$) for FIR mixing experiments. The measurements indicate, however, that the modulation depth can be large ($\geq 50\%$); hence that samples grown by Prof. Witt's technique may ultimately be suitable for phase-matched, FIR difference frequency generation.

F. Excitation of Collective Modes in Narrow Gap Semiconductors

Two mechanisms for exciting plasma waves in semiconductors have been studied theoretically. The first, involving nonlinear excitation by two laser beams, has the advantage of exciting plasmons of well defined wave vector. Their presence can be detected by radiation they emit at the plasma frequency. An experiment to test this idea in crystals of n-InSb is now being set up.

The second plasmon excitation mechanism under consideration requires optical pumping of semiconductor crystals [such as (Pb,Sn)Te or (Cd,Hg)Te] whose bandgap can be made equal to their collective mode energy. When this condition is satisfied; minority carriers recombine via plasma wave emission. The process is exceedingly efficient, and reduces carrier lifetimes to the picosecond range. The reduction in lifetime is an interesting effect with several potential applications. However, the plasmon emission process could be even more important if means were found to channel the energy into a small subset of the plasma modes. Under such conditions, stimulated plasma wave emission becomes a realistic possibility. Calculations are in progress to test several schemes, including finite geometry and the application of magnetic field, to control the plasmon spectrum, and thus permit selective excitation of long wave length plasmons.

G. Studies of the Gunn Effect via Four-Photon Mixing

Calculations imply that four-photon mixing by electrons in n-GaAs is greatly enhanced in the Gunn effect regime. A preliminary

experiment by Prof. Muehlner has confirmed this suggestion.

Work on this problem has been temporally discontinued due to Muehlner's departure. However, these studies will be resumed in the near future.

III. Publications

- "Theory of Spin-Flip Line Shape in CdS," P.A. Wolff, J.G. Ramos and S. Yuen, Proceedings of 1st Soviet-American Symposium on the Theory of Light Scattering in Solids (Publishing House "Nauka", Moscow, 1976).
- "Spin-Flip Nonlinearities in Semiconductors," P.A. Wolff, in Laser Photochemistry Tunable Lasers and Other Topics (Addison-Wesley, Reading, Mass. 1976).
- "Spin Dynamics and Four-Photon Mixing in InSb," V.T. Nguyen, E.G. Burkhardt, and P.A. Wolff, Optics Comms. 16, 145 (1976).
- "Coherent Effects in Spin-Flip Light Scattering," P.A. Wolff, R.N. Nucho, and R.L. Aggarwal, IEEE Jour. Quant. Elects. QE-12, 500 (1976).
- "Dispersion of the Nonlinear Optical Susceptibility of n-Type Germanium," R.A. Wood, M.A. Khan, P.A. Wolff and R.L. Aggarwal, Optics Comms. 21, 154 (1977).
- "Free-Electron Nonlinear Optical Processes in Semiconductors," P.A. Wolff, Proceedings of the 16th Scottish Universities Summer School, (Academic Press, New York, 1977).
- "A Chemical Bond Approach to the Electric Susceptibility of Semiconductors, I," R.N. Nucho, J.G. Ramos and P.A. Wolff, Phys. Rev. B 17, 1843 (1978).
- "A Chemical Bond Approach to the Electric Susceptibility of Semiconductors, II," R.N. Nucho, J.G. Ramos and P.A. Wolff, Phys. Rev. B 17, 4835 (1978).
- "Resonant Four Wave Mixing in n-Type Silicon," M.A. Khan, D.J. Muehlner, and P.A. Wolff, Optics Comms. 30, 206 (1979).

IV. Personnel

P.A. Wolff, Prof. Physics

D.J. Muehlner, Asst. Prof. Physics

Andrew Wood, Sponsored Research Staff

Jose Ramos, Visiting Scientist

Graduate Students:

Roger Nucho: Research Assistant

Lynn Detwiler: Bell Fellow

Roosevelt People: Bell Fellow

Asif Khan: Research Assistant

Kathy Kash: Research Assistant

V. Degrees Granted

"Thin Film Plasmons as a Nonlinear Mixing Mechanism for Difference Frequency Generation," L. Detwiler, M.S., E.E., February 1976.

"Chemical Bond Approach to the Electric Susceptibility of Semiconductors," R.N. Nucho, Ph.D., Physics, June 1977.

"Four-Photon Mixing in Semiconductors," M.A. Khan, Ph.D., Physics, May 1979.

VI Interactions

A. Bell Laboratories

There is an ongoing interaction with Bell Laboratories via a consulting arrangement; Prof. Wolff visits Bell for a few days each month. He has regularly discussed nonlinear optics problems with Drs. Bridges, Geschwind, Gornik and Nguyen there. The interaction with T.J. Bridges has been especially fruitful. Bridges has performed four-photon mixing experiments, similar to those done at MIT, in (Cd,Hg)Te crystals grown by Cominco.

B. National Magnet Laboratory

We collaborate with Dr. Roshan Aggarwal of the National Magnet Laboratory. He participated, and was a co-author, in the four-photon mixing work in Ge, and in our attempts to demonstrate impurity-Raman scattering. We consider Dr. Aggarwal a member of the IR research group, and meet with him regularly (typically, twice a month). In addition, Dr. Aggarwal is supervising the experimental work of Roosevelt People. Prof. Margaret Weiler (formerly of NML) also meets with the group. She is an expert on the properties of (Cd,Hg)Te.

C. Honeywell

Dr. Paul Kruse has provided us with a large, excellent quality (Cd,Hg)Te crystal for use in nonlinear optic experiments. We expect, via Honeywell's AFOSR project, to receive a steady supply of such material. Dr. Kruse and

his associates have visited our laboratory several times in recent years. In addition, as indicated above, Asif Khan spent the summer of 1978 at Honeywell and is now a staff member there. He will facilitate future contacts with Honeywell.

D. Prof. Witt, Materials Science, MIT

Prof. Witt is preparing (with AFOSR support) n-InSb crystals with layered doping for our nonlinear optics work. We have already received some heavily doped material from him. There is continual interaction between his students and ours.

E. Prof. Fielding Brown, Williams College

Prof. Brown spent the past year on sabbatical at MIT. He assisted us in setting up FIR lasers for use in stimulated impurity Raman scattering experiments, and generally advises us concerning the problems of FIR laser operation.

F. Dr. Aram Mooradian, Lincoln Laboratories

We have occasional contact with Drs. Brueck and Mooradian. Dr. Mooradian provided the $(\text{CdGe})\text{As}_2$ crystal for doubling our CO_2 lasers.

G. Prof. Michael Salour, MIT

Several members of Salour's group regularly attend our group meetings. They, too, are interested in nonlinear optical effects in semiconductors.

VII. Patents

None.