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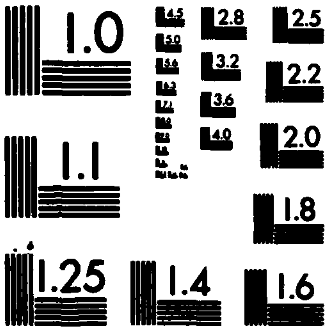

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

This program, which combined theory and experiment, has used nonlinear optic techniques to study electron dynamics in semiconductors. Possible applications were also considered. Areas of investigation included:

1) Donor structure in Si and Ge. Four-wave mixing experiments were used to determine donor energies, radii, diamagnetism, g-values, and deformation potentials.

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ii) Infrared radiation via collective modes. Coherently excited plasma waves, in n-InSb plates, radiate at 100 μ.

iii) Acoustic plasmons. Light scattering experiments, in optically excited electron-hole plasmas, gave the first evidence for acoustic plasma waves in solids.

iv) Theoretical calculations indicate that stimulated plasma wave emission can be achieved in a semiconductor with $\hbar\omega_p = E_G$. The optimal material is $(Hg_{1-x}Cd_x)Te$, with $x < 0.12$, and field-tunable gap induced by a sizable ($\simeq 50$ kG) magnetic field.

v) FIR difference frequency generation. Uniaxial strain has been used to enhance FIR (100 μ) power in n-InSb.

vi) Measurement of picosecond relaxation times. The free-carrier-induced nonlinear susceptibility of semiconductors has been shown to have a strong difference frequency dependence at small $\Delta\omega$. These experiments determine electron thermalization times and valley transfer times, in the picosecond range, for n-HgCdTe, n-GaSb, n-InSb and n-GaAs.

vii) Spin-induced four-wave mixing has been used to accurately characterize $Hg_{1-x}Cd_xTe$ crystals.

viii) Several novel mechanisms for large, fast $\chi^{(3)}$'s have been explored.

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I. Research Objectives

This is the final technical report for AFOSR Contract F49620-80-C-0008, "Infrared Nonlinear Processes in Semiconductors." The primary purpose of ~~the~~ this program was to use nonlinear techniques to study electron dynamics and kinetics in semiconductors. Possible applications of nonlinear interactions were also considered. These investigations have combined theory and experiment.

The statement of work for the contract included the following tasks:

A) Study impurity dynamics and electron delocalization in Ge and Si;

B) Generate infrared radiation via coherent excitation of collective modes;

C) Observe and study magnetoacoustic waves;

D) Stimulate plasmon emission in (Hg,Cd)Te;

E) Study spin dynamics and use spins to generate far infrared radiation in (Hg,Cd)Te;

F) Study valley-transfer processes;

G) Characterize (Hg,Cd)Te with optical techniques; and

H) Study semiconductors with field-tunable gap.

Accomplishments in these areas are described below. Work concerning large infrared nonlinear coefficients is also summarized.

II. Research Projects

A. Nonlinear Optical Impurity Interactions in Ge and Si

The resonant, four-wave mixing technique was used to study[3,4,5,17] the magnetic field dependence of the valley-orbit splitting, 4Δ, between the singlet, $1s(A_1)$, and tripley degenerate, $1s(T_2)$, levels of shallow donors in Ge. Experiments were performed with B along [100] and [111] directions. In the $B \parallel [100]$ case, the diamagnetic shift determines the radius of the ground state wave function. For $B \parallel [111]$, the $1s(T_2)$ level splits into two levels, one of "high" and the other of "low" diamagnetism. The diamagnetism and g-factor ($g \simeq 1.07$) of the high diamagnetism level were measured to $B = 70$ kG. Transitions of both spin-up and spin-down electrons were observed, indicating that spins are heated during the 100 nsec laser pulses. Excellent agreement was found between measured values of 4Δ vs. B and those calculated by Larsen, including magnetic field compression of the wave function.

A detailed theory of the field-induced splitting of the $1s(A_1)$ and $1s(T_2)$ levels was developed.[9] These calculations imply level anti-crossing effects for $B \simeq 140$ kG.

Four-wave mixing spectroscopy has also been used to study[23] the stress dependence of the ground state multiplet of phosphorus donors in Si. This work required development of a calibrated stress cryostat. The experiments determine the stress deformation potential constant, Ξ_u, of the 1s ground state multiplet.

Finally, in Ge:As, four-wave experiments under stress and field, have been used to study valley repopulation and magnetic compression effects on the ground state, $1s(A_1)$, wave function. The measured diamagnetic coefficient for the $1s(A_1) \rightarrow 1s(B_1)$ transition is in excellent agreement with the theoretical value.

B. FIR Generation by Coherently Excited Collective Modes

Coherent far infrared radiation, in the 100 μ wavelength range, was generated[15,19] by plasma waves excited in thin n-InSb plates. The plasma waves were nonlinearly excited, via the plasmon Raman interaction, by two CO_2 lasers at frequencies ω_1 and ω_2. Radiation was only produced when the difference frequency, $\Delta\omega = \omega_1 - \omega_2$, was near the plasma frequency, ω_p. In a thin film, two surface plasmons couple to form a simple, partially transverse mode, which radiates efficiently. A thin-film plasmon mode of definite wave vector was selectively excited by a longitudinal driving force at $\Delta\omega$, generated by two noncolinear laser beams. The plasmon wave vector in the plane of the film is controlled by the angle θ between the laser beams.

Signals observed in these experiments were somewhat weaker than anticipated, but otherwise demonstrated all features predicted by our theory of the nonlinear excitation process. Some work has also been done on (Hg,Cd)Te epilayers where bandgap resonance of CO_2 laser pump beams enhances the excitation. Unfortunately, these layers did not have plasma frequencies in the accessible $\Delta\omega$ range. However, far infrared signals due to coherent phonon emission were seen. To our knowledge, this experiment is the first observation of phonons with CO_2 lasers.

C. Acoustic and Magnetoacoustic Plasma Waves

Though acoustic plasma waves were predicted many years ago, they have not been observed in crystals until recently. Light scattering spectra of optically excited electron-hole plasmas in GaAs were shown to exhibit a novel, low frequency collective resonance.[16] The position and carrier - density variation of this line agree with those predicted by the RPA theory of the acoustic plasmon. In previous light scattering studies of optically excited electron - hole gases, the acoustic plasmon resonance was not observed, presumably because of plasma inhomogeneity. Uniform plasmas were assured in this work by creating electron - hole pairs in thin (≈ 4000 Å) GaAs eiplayers, bounded by transparent (Ga,Aℓ)As layers.

A theoretical study of magnetoacoustic wave linewidths suggests that those excitations should be observable in such materials as n-InSb or n-PbTe. The experiments are far from easy, however, and have been postponed in favor of more pressing and rewarding work.

D. Plasmon Excitation and Emission in (Hg,Cd)Te

Theoretical calculations[10,11] indicate that is possible to stimulate plasma wave emission in a narrow-gap semiconductor, subjected to a large magnetic field, when the plasma frequency ω_p is tuned to the bandgap E_G of the semiconductor. In the absence of a magnetic field, minority carriers are believed to recombine via plasma wave emission when $E_G \leqslant \hbar\omega_p$, with lifetimes in the $10^{-12} - 10^{-13}$ sec range. This rapid recombination precludes stimulated plasma wave emission.

Application of a magnetic field changes the plasmon recombination process in two important ways:

(1) Landau level quantization enhances the electron and hole densities-of-states at the band edges. This enhancement increases the gain of long wavelength, $k = 0$, plasmon modes.

(2) The magnetic field modifies the plasmon dispersion relaxation. This effect can be exploited to inhibit recombination via plasmon modes propagating across the field, resulting in a substantial increase in the recombination time. These two effects combine to yield a threshold pump

level for stimulated plasma wave emission in the 10 - 100 kW/cm^2 range, which is modest by CO_2 laser standards.

The theory suggests that stimulated plasma wave emission will be most easily achieved in a material such as $Hg_{1-x}Cd_xTe$, with $x < 0.12$. The alloy is a semimetal (or zero gap semiconductor) in the composition range $0 < x \lesssim 0.15$. Application of a magnetic field produces a small field, tunable gap which can readily be matched to the plasma frequency. Other advantages of $Hg_{1-x}Cd_xTe$ relative to the Pb-salt alloys include:

(1) HgCdTe can easily be doped to the concentration range desired.

(2) In the zero gap range, the bandgap and effective mass can be varied independently of one another. This fact is helpful in optimizing the gain.

To calculate the gain of plasma waves when $E_G \simeq \hbar\omega_p$, we have extended Kubo's analysis, of the density-of-states in a large magnetic field, to the interband, joint-density-of-states problem.[13] Ladder diagrams describing scattering by correlated electron - hole pairs were summed and evaluated. These ladder diagrams cause a sizable modification of the interband matrix element.

The plasma wave instability is best studied by exciting it in thin plates. There should then be substantial radiation at frequency ω_p. Measurements of this type require high quality $Hg_{1-x}Cd_xTe$ epilayers with appropriate composition and doping level. To date, experiments have been precluded by our inability to obtain good layers. However, with the development of an in-house crystal growth facility, they will be available in the continuing phase of this program.

E. Spin-Induced FIR Generation

FIR generation via spin-induced difference frequency mixing was first observed in n-InSb some years ago. The FIR power in these experiments was modest (10 μW); nevertheless, there was considerable interest in the system as a potential source of magnetically tunable FIR radiation.

Recently, Kuchar et al. have demonstrated that the application of a uniaxial stress to n-InSb dramatically enhances the spin resonance transition. To make use of this effect, we have developed a calibrated, uniaxial stress cell. The stress cell has been coupled to a superconducting magnet and TEA CO_2 laser system to demonstrate enhancement of the FIR generation. An enhancement factor of 30 (in FIR power) was observed for stress F = 2 kbar applied parallel to the direction of magnetic field along a [111] crystal direction. There is also a marked change (13% at 2 kbar) in the electron spin resonance frequency. Unfortunately, the enhancement is accompanied by severe line broadening. This fact, and difficulties in phase matching, will preclude the use of strain enhancement for tunable FIR generation.

F. Study of Valley-Transfer Times and Other Fast Electronic Relaxation Times via Four-Wave Mixing

A variety of experiments have been performed to demonstrate that four-wave mixing provides a new, relatively simple technique for measurement of fast relaxation times in semiconductors. In the first measurements[12] the free-carrier-induced $\chi^{(3)}$ of n-InSb was studied as a function of the difference frequency, $\Delta\omega$, between two CO_2 laser beams. The four-wave signal is enhanced by a factor of 100 as $\Delta\omega$ becomes small; the effect has since been seen in a number of other semiconductors. We explain it by a model which requires two distinct perturbations of the electron distribution — an oscillation in velocity space, and a temperature fluctuation. Both contribute to $\chi^{(3)}$; the contribution of temperature fluctuations varies rapidly with $\Delta\omega$. The theory gives an excellent fit to the data. Energy relaxation times of 3 - 4 psec are inferred for the highly excited electron gas in the n-InSb crystal.

These experiments were then continued[21] in n-GaAs and n-GaSb. Large frequency ($\Delta\omega$) and temperature variations in $\chi^{(3)}$ were observed in both cases. At lower temperatures, the results are explained by the temperature fluctuation model developed for the n-InSb case. The fit to the model determines the energy relaxation time τ_E of carriers. For example, in n-GaAs at room temperature $\tau_E = 0.7$ psec. In n-GaSb at 90°K, $\tau_E \simeq 10$ psec.

As temperature is increased (to 800°K in n-GaAs or 300°K in n-GaSb), carriers transfer to subsidiary minima, at the L-points in the Brillouin zone. Valley transfer modifies the $\chi^{(3)}$ vs. $\Delta\omega$ variation. Transport theory was used to derive formulas for the drift momentum, temperature, and valley populations of the electron system under such conditions. These equations, in turn, enable one to calculate $\chi^{(3)}$ vs. $\Delta\omega$ and T. The theory gives a good fit to the data, and determines both electron thermalization and valley - transfer times. For n-GaAs at 800°K, $\tau_E = 2 \times 10^{-13}$ sec, and the valley - transfer time is about 1 psec. The valley - transfer deformation potential inferred from this result agrees with that previously estimated.

Four-wave mixing has also been used[18] to study the variation of $\chi^{(3)}$ with difference $\Delta\omega$ and laser intensity I in low carrier concentration HgCdTe crystals. At small $\Delta\omega$, $\chi^{(3)}$ is caused by nonparabolicity of free electrons generated by two-photon absorption, with $\chi^{(3)}$ scaling as $(\Delta\omega)^{-2}$ and $I^{2/3}$. The $\Delta\omega$ variation of $\chi^{(3)}$ indicates that the electron thermalization time is longer than 8 psec. At large $\Delta\omega$, $\chi^{(3)} \simeq 3 \times 10^{-8}$ esu is mainly due to bound electrons.

Picosecond relaxation times are not easy to measure directly. Thus, we believe that nonlinear optic studies of the frequency variation of $\chi^{(3)}$ may provide an important, general, quasi-CW method for determining fast relaxation times in semiconductors.

G. Characterize (Hg,Cd)Te with Optical Techniques

IR absorption[6] and spin-induced four-wave mixing[2] were used to characterize $Hg_{1-x}Cd_xTe$ crystals. The four-wave technique is particularly sensitive. Spin resonance lines with widths of 10 G (out of 2000 G) were observed in optimal cases. Such sharp resonances enable one to estimate the local x-value to .01%. At other points on the crystals, several resonances were observed, suggesting that the material contains substantial inhomogeneities due, presumably, to constitutional supercooling during growth.

H. Study Semiconductors with Field - Tunable Gap

In semimetallic $Hg_{1-x}Cd_xTe$ $(x < 0.15)$, a magnetic field creates a small energy gap, typically 30 meV at 50 kG. Attempts were made, with a FIR laser, to observe this field-induced gap via direct optical absorption. After extensive efforts it was concluded that sample quality was not sufficiently good to provide a meaningful test of the idea. This work will continue, in the new contract, with $Hg_{1-x}Cd_xTe$ or $Hg_{1-x}Mn_xTe$ epilayers.

J. Large Infrared Nonlinear Coefficients

Several processes giving rise to large $\chi^{(3)}$'s have been identified. Theory implies that in semiconductor crystals with spatially varying effective mass, there is a large enhancement of the free - carrier nonlinear susceptibility. Structures of this type can be grown via MBE techniques. The calculations show that the variable-mass effect is most pronounced when the mass varies rapidly in space, though large amplitude fluctuations are not required. Nonlinear coefficients in the 10^{-4} - 10^{-5} esu range are anticipated at 10.6 μ.

Intense, degenerate four-wave mixing has been observed[24] at room temperature in p-type HgCdTe with a CO_2 laser pump. The experiments yield a nonlinear susceptibility, $\chi^{(3)} = 5 \times 10^{-6}$ esu, with $p = 2.5 \times 10^{17}$ holes/cc. This value exceeds, by several orders of magnitude, any previously observed nonresonant, nonlinear susceptibility. The large $\chi^{(3)}$ is attributed to intervalence band transitions which modulate the plasma contribution to the dielectric function. The nonlinear susceptibility is proportional to the light-to-heavy hole relaxation time, which was estimated to be $2\text{-}4 \times 10^{-12}$ sec from these measurements.

III. Publications

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IV. Theses

1. "Resonant Nonlinear — Optical Studies of Magnetically Tunable Donor Impurity States in Germanium," Roosevelt People, Ph.D. Thesis in Physics, MIT, September 1980.

2. "Nonlinear Optical Studies of Electron Dynamics in Semiconductors," Kathleen Kash, Ph.D. Thesis in Physics, MIT, September 1982.

3. "Radiation from Coherently Excited Plasmon and Phonon Modes in Semiconductor Thin Films," John Barry McManus, Ph.D. Thesis in Physics, MIT, September 1982.

V. Professional Personnel

Prof. P.A. Wolff, Principal Investigator

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R. People, Physics Graduate Student

K. Kash, Physics Graduate Student

J.B. McManus, Physics Graduate Student

E. Youngdale, Physics Graduate Student

E. Gamble, EE&CS Graduate Student (GE Fellow)

J. Warnock, Physics Graduate Student (Bell Fellow)

G. Boebinger, Physics Graduate Student (Hertz Fellow)

S. Wong, Physics Graduate Student

VI. Interactions

Drs. Aggarwal and Jagannath have collaborated, throughout this program, with Dr. David M. Larsen of the Francis Bitter National Magnet Laboratory. Dr. Larsen's theoretical assistance has been invaluable in interpreting the four-wave mixing experiments on donors in Si and Ge.

Interactions with Dr. Paul Kruse's group at the Honeywell Corporate Research Center, Bloomington, MN, and with Dr. James Shanley's group at the Honeywell Electro-Optics Center, Lexington, MA continue. In particular, these groups have provided us with a number of (Hg,Cd)Te crystals and epilayers.

Prof. Wolff interacts regularly with the Bell Laboratories optics group through a consulting arrangement. The work on acoustic plasmons, described under item C above, was performed in collaboration with Drs. Pinczuk and Shah of that group.

Our group seminars have often been attended by Profs. Haus and Ippen, and members of their groups, who are in the M.I.T. EE&CS Dept. Haus and Ippen have an interest in picosecond optical signal processing; discussions with them stimulated much of the work described under item J above.

Finally, Dr. Aggarwal is an active member of the Program Committee for U.S. Workshops on the Physics and Chemistry of Mercury Cadmium Telluride. The last workshop took place February 8-10, 1983 in Dallas; the next will be held May 15-17, 1984 in San Diego. These workshops are supported by DARPA and ARO.

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