Intervalance band relaxation times have been measured, in the picosecond range, in p-type GaAs and Ge. These measured times are in agreement with those calculated for optic phonon mediated processes. An anticrossing, predicted by People and Wolff, has been observed between As donor levels of opposite spin in Ge. A theory has been developed, which, with no adjustable parameters, provides good agreement with experiment.
I. Summary of Research Goals

The following research topics are planned. Most of them involve nonlinear optical processes in semiconductors. The nonlinear interactions are used to probe the electron and hole dynamics in semiconductors.

a.) Measurement of picosecond relaxation times in p-type semiconductors, using four-wave mixing of CO$_2$ laser beams.

b.) Observation of anticrossing between donor levels of opposite spins in germanium, predicted by People and Wolff, using four-wave magnetospectroscopy.

c.) Study of the spin-flip Raman linewidth in n-type InSb, using four-wave mixing with CO$_2$ lasers.

d.) Observation of magnetoacoustic waves in n-type InSb, via Raman gain experiments.

e.) Attempt stimulated plasma wave emission in n-type Hg$_{1-x}$Cd$_x$Te with $x = 0.24$, via Raman gain experiments.

f.) Study third-order nonlinear susceptibility $\chi(3)$ in uniaxially stressed p-type semiconductors Si, Ge, GaAs, Hg$_{1-x}$Cd$_x$Te, using four-wave mixing with CO$_2$ laser beams.
II. Current Status of the Research Effort

The research project uses nonlinear optical processes to study electron dynamics in semiconductors. Significant progress has been made in the following areas:

(i) Study of the Intervalance Band Relaxation Times via Four-Wave Mixing with CO₂ Lasers

(Item a. above)

Free carriers in semiconductors cause large third-order optical nonlinearity $\chi^{(3)}$ which can be measured via four-wave mixing. Dispersion of $\chi^{(3)}$ has previously been used for a measurement of the electron energy relaxation time and valley transfer time, in the picosecond regime, in n-type semiconductors InSb, GaAs, and GaSb. Hole-induced $\chi^{(3)}$ has been studied in p-type semiconductors for the first time. Measured values of $\chi^{(3)}$ in p-type GaAs and Ge are found to be in agreement with those predicted from the intervalence band population modulation mechanism. Dispersion of $\chi^{(3)}$ yields intervalence band relaxation times in the pico-second regime. These measured times are in agreement with those calculated for optic phonon mediated processes. Such information is useful in the design of devices employing intervalence band transitions. In particular, our work suggests that the optimal material for such applications should be nonpolar and have a small bandgap.
(ii) Four-Wave Spectroscopy of Donors in Semiconductors (Item b. above)

Four-wave spectroscopy with CO$_2$ lasers has been used to study Raman allowed transitions between the valley-orbit split $1s$ states of As donors in Ge at 1.8K, using magnetic field $B$ up to 12T. With $\hat{B} \parallel [111]$ the $1s(T_2)$ state splits into six levels (including spin), two with high and four with low diamagnetism. An anticrossing, predicted by People and Wolff [Phys. Rev. B24, 4634 (1981)] has been observed between donor levels of opposite spin. Valley repopulation due to breaking of the tetrahedral symmetry by the magnetic field is also observed. A theory has been developed, which, with no adjustable parameters, provides good agreement with experiment.

(iii) Study of the Spin-flip Raman Linewidth in n-type InSb (Item c. above)

The lineshape for the spin-flip transition in n-type InSb has been investigated via four-wave spectroscopy with CO$_2$ lasers. In contrast to lineshapes obtained previously with pump laser frequencies close to the bandgap, the lineshape in our experiment shows a pronounced asymmetry. Furthermore, an additional peak is resolved at low laser powers. The asymmetric lineshape cannot be understood in terms of magnetic joint-density-of-states alone.
(iv) Stimulated Plasma Wave Emission in n-type Hg$_{1-x}$Cd$_x$Te (item e. above)

Our calculations indicate a plasmon Raman gain of 10 cm$^{-1}$ with peak pump power of 100 kW/cm$^2$ for a Hg$_{1-x}$Cd$_x$Te crystal with $x = 0.24$ (to match the bandgap at $T = 4K$ to the CO$_2$ laser energy), and $n = 3 \times 10^{15}$/cm$^3$.

We plan to obtain high homogeneity samples from Honeywell Electro-Optics Operation, Lexington, MA. The concept would be tested via Raman gain experiments using counterpropagating CO$_2$ pump (TEA laser) and probe (cw laser) beams.
III. Publications


5. E.R. Youngdale, D.M. Larsen and R.L. Aggarwal, "Observation of Anti-Crossing between Zeeman-split 1s(T$_2$) States of Opposite Spin for As Donors in Germanium" to be submitted to Phys. Rev. B.

IV. Professional Personnel

Prof. P.A. Wolff, Principal Investigator

Dr. R.L. Aggarwal, Co-Principal Investigator

Dr. D.M. Larsen, Research Scientist

Dr. C. Jagannath, Research Associate

Prof. L.R. Ram-Mohan, Consultant

Dr. S.Y. Yuen, Research Scientist

E. Isaacs, Physics Graduate Student

J. Warnock, Physics Graduate Student

S. Wong, Physics Graduate Student

E.R. Youngdale, Physics Graduate Student
V. Interactions

We are continuing our interaction with the Honeywell Research Group (Bloomington, MN) through Dr. Paul Kruse and Honeywell Electro-Optics Operation through Dr. Marion Reine. We expect to obtain high homogeneity n-type samples of Hg$_{1-x}$Cd$_x$Te (with $x = 0.24$ and $n_e = 3 \times 10^{15}$ cm$^{-3}$), grown by Dr. Don Nelson at the Honeywell Electro-Optics Operation, for stimulated plasma wave emission experiments.

We have an ongoing interaction with AT&T Bell Laboratories through Prof. Wolff's consultancy. Dr. R.A. Logan has provided p-type GaAs samples used for the measurement of picosecond intervalence band relaxation times.

At MIT we strongly interact with Dr. P. Becla and Prof. August Witt's and Prof. H. Gatos' groups regarding growth and characterization of materials. We also interact with Prof. E. Ippen and Prof. H. Haus with regard to nonlinear optical device applications.