10. Infrared Nonlinear Optics

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10.1 Infrared Nonlinear Processes in Semiconductors

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In doped, narrow gap semiconductors there is a sizeable nonlinear (Raman) interaction between two laser beams and plasma waves. This interaction can be used to coherently excite plasmons of well-defined wave vector. An experiment to test this idea was performed[1] in thin n-InSb plates; plasmon excitation was detected via FIR radiation ($\simeq 100\mu$) emitted at the plasma frequency by the thin-film modes. The FIR power observed was lower than anticipated, but in other respects the measurements confirmed theory.

Though acoustic plasma waves were predicted many years ago, they have not been observed in crystals until recently.[2] Light scattering spectra of optically excited electron-hole plasmas in GaAs exhibit a novel, low frequency resonance. The position and carrier-density variation of this line agree with those predicted by 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 ($\simeq$ 4000 A) GaAs epilayers, bounded by transparent (Ga,Al)As layers. The investigation was performed in collaboration with Dr. A. Pinczuk and Dr. J. Shah of Bell Laboratories.

Intense, degenerate four wave mixing has been observed[3] at room temperature in p-type (Hg,Cd)Te 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 is estimated to be (2 – 4) $\times 10^{-12}$ sec.

Theoretical calculations[4] suggest 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 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.

Nonlinear optical studies of donors in Ge and Si are continuing. Four-wave mixing spectroscopy has been used to study[5] the stress dependence of the ground state multiplet of phosphorus donors in Si. This work was made possible through the development of a quantitative stress cryostat. The experiments determine the stress deformation potential, Ξ_a, characterizing the 1s ground state multiplet.

The stress cell has also been coupled to a superconducting magnet to measure the combined stress (F) and magnetic field (B) dependence of the donor levels. With F constant one observes four-wave resonances, as a function of B, for various values of the difference frequency, $\Delta\omega$, between two CO_2 laser beams. Preliminary experiments on As donors in Ge with stress F || [110] yield a value 0.03 cm^{-1}/T^2 for the diamagnetic coefficient of the 1s $(A_1) \rightarrow 1s(B_1)$ transition in the high stress limit. This value is in excellent agreement with a theory which takes into account both valley re-population and the change in size of the ground state, $1s(A_1)$, wave function.

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