

## 9. Infrared Nonlinear Optics

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## 9.1 Infrared Nonlinear Processes in Semiconductors

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The difference-frequency variation of the free-carrier-induced, third-order nonlinear susceptibility [ $\chi^3$ ] has been studied in n-InSb, n-GaAs, n-GaSb, and n-(Hg,Cd)Te. Two CO<sub>2</sub> laser beams (of frequencies  $\omega_1$  and  $\omega_2$ ) were focused onto the samples. The four-wave signal, at frequency  $\omega_3 = 2\omega_1 - \omega_2$ , was measured as a function of  $\Delta\omega \equiv (\omega_1 - \omega_2)$  in the range  $1.5 \text{ cm}^{-1} \leq \Delta\omega \leq 100 \text{ cm}^{-1}$ . A dramatic enhancement of  $P(\omega_3)$ , as  $\Delta\omega \rightarrow 0$ , was seen in all cases. This effect had not previously been observed, nor was it predicted by theories of free-carrier-induced, four-wave mixing. In n-InSb, and in n-GaAs and n-GaSb at low temperatures, we attribute the effect<sup>1</sup> to temperature fluctuations of the electron gas in the crystal. The model explains the  $\Delta\omega$  variation of  $\chi^3$  and yields values for the electron-lattice thermalization times. At higher temperatures, valley-transfer processes contribute to the nonlinear susceptibility<sup>2</sup> in n-GaAs and n-GaSb, modifying the  $\Delta\omega$  variation of  $\chi^3$ . We have developed coupled equations for the temperature and carrier-density fluctuations, which give an excellent fit to the data, and yield values for both the electron thermalization and valley-transfer times. Finally, in (Hg,Cd)Te, the four-wave signal is attributed<sup>3</sup> to the electron-hole plasma generated by the intense CO<sub>2</sub> laser beams. This work suggests that four-wave mixing may prove to be a versatile technique for studying short electron-relaxation times (in the  $10^{-12}$  sec range) in semiconductors.

Nonlinear optical studies of donors<sup>4</sup> in n-Ge are continuing. Four-wave mixing has been used to study the magnetic field dependence of the valley-orbit splitting for B || [111] in Ge:P and Ge:As. In this geometry, the valley-orbit split manifold contains a singlet (with large diamagnetism) and a doublet (with small diamagnetism). We observe transitions, for electrons of both spin directions, from the ground state to the singlet. The data determine the donor diamagnetism (hence the ground-state radius) and the g-value of the excited state, and give information concerning the magnetic field compression of the wave functions.

Far infrared generation via coherent excitation of thin-film plasmons has been observed in n-InSb<sup>5</sup>. Thin plates of the semiconductor were irradiated with two CO<sub>2</sub> laser beams whose difference frequency,  $\Delta\omega = (\omega_1 - \omega_2)$ , was near the plasma frequency ( $\omega_p$ ) of the electron gas in the crystal. Plasma waves, at frequency  $\Delta\omega$ , were coherently excited by the nonlinear interaction responsible for plasmon-Raman scattering. The wave vector of the waves was controlled by varying the angle between the laser beams. Plasma waves in thin plates radiate efficiently at an angle determined by their wave vector. The observed far infrared signals behave, in all respects, as predicted by the theory of coherent plasma-wave excitation. Powers in the 100-microwatt range are measured. This power can be enhanced via band-gap resonance of the pump lasers.

#### References

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