7. Infrared Nonlinear Optics

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7.1 Four Wave Spectroscopy of Donors

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, using magnetic field to 12 T. With B ‖ [111] the 1s (T_2) state splits into six levels (including spin), two with high diamagnetism and four with low diamagnetism. An anti–crossing predicted by People and Wolff[1] was observed between donor levels of opposite spin. A theory has been developed that, with no adjustable parameters, provides good agreement with experiment. This work determines electron g–values, valley repopulations, and magnetic wave function compressions.[2] Overall, it constitutes a searching test of donor theory.

7.2 Stressed–Induced Far Infrared Generation

FIR generation via spin–induced difference frequency mixing was first demonstrated in n–InSb some years ago, but the FIR powers were modest ($\approx$ 10 μW). More recently, Kriechbaum *et al.*[3] showed that uniaxial stress dramatically enhances spin resonance absorption in n–InSb. Spin–induced FIR emission should also be enhanced. We have performed experiments to test this idea; an enhancement factor of 30, in FIR power, was observed with a 2 kbar uniaxial stress. In addition, there was a marked change ($\approx$ 13% at 2 kbar) in the spin resonance frequency.[4] Unfortunately, the enhancement is accompanied by severe line broadening. This fact, and difficulties in phase matching, will probably preclude the use of strain enhancement for tunable FIR generation.

7.3 Intervalence Band Relaxation Times

L.R. Ram-Mohan, Peter A. Wolff, Sunny Y.C. Yuen

Free carriers in semiconductors give rise to large, third order optical nonlinearities that can be studied via four-wave mixing. In our previous work, the dispersion of $\chi^{(3)}$ was used to determine electron energy relaxation times and valley transfer times in n–InSb, n–GaAs, and n–GaSb. More recently we have investigated hole-induced optical nonlinearities in p–GaAs and p–Ge.[5] Large ($\approx 10^{-7}$ esu) $\chi^{(3)}$'s are observed; they are attributed to intervalence band population modulation. The dispersion (variation with $\triangle \omega = \omega_1 - \omega_2$) of $\chi^{(3)}$ determines the intervalence band relaxation times of the holes, which are in the 3×10^{-12}–10^{-13} sec. range. The measured relaxation times are in agreement with those calculated from first principles for optic phonon emission. This work confirms our mechanism for hole-induced optical nonlinearities, and suggests that the optimal material for devices employing intervalence band transitions should be nonpolar and have a small band gap.

7.4 Nonlinear Optics Near the Metal–Insulator Transition

Peter A. Wolff, Sunny Y.C. Yuen

Free carriers in semiconductors cause optical nonlinearity whenever the properties of the electron states vary with energy. The metal–insulator transition is an extreme example of such a situation since the Fermi level then separates localized from delocalized states. This idea is confirmed by experiment; our four wave mixing measurements have shown that n–Si:P has very large nonlinear coefficients (per carrier) for doping levels near the transition (n $\approx 3.7 \times 10^{18}$/cc), but small ones in lightly doped crystals.[6] The faster than linear variation of $\chi^{(3)}$ with n implies that electron-impurity interactions are responsible for the nonlinearity. To our knowledge, this is the first example of a sizable optical nonlinearity caused by scattering.

We have developed a theory of this effect which shows that it provides a direct measure of the electron pile–up at impurity sites.[6] Experimentally, this quantity varies five times more rapidly (with energy) than RPA predicts. We attribute the discrepancy to higher order terms in the perturbation theory, which cause carrier diffusion and localization.

The theory also implies that the nonlinearity is sensitive to dopant type. Further studies, with Si crystals doped and alloyed in alternate ways will be performed to optimize the nonlinearity.

7.5 Nonlinear Optics in Superlattices

Sunny Y.C. Yuen

Yuen has suggested several mechanisms for enhancing free–carrier–induced nonlinear optical processes in superlattices.[7] Initially this work was entirely theoretical. Recently, however, structures for testing these ideas have been made available to us. In particular, we have recently received the following superlattices:

1. n–GaAs/(GaA1)As with quantum level spacing adjusted to match the CO_2 laser energy

2. p–InGaAs/GaAs strained layer superlattice

3. InSb/(InAl)Sb

Preliminary four–wave mixing experiments have been performed on all of these samples. The InSb/(InAl)Sb superlattices give large signals ($\chi^{(3)} \approx 10^{-7}$ esu) — despite their thinness — that we attribute to intervalence band population modulation. These experiments will continue with better characterized InSb/(InAl)Sb superlattices and InSb/In(AsSb) superlattices.

The p–(InGa)As/GaAs SLS has only a modest nonlinear susceptibility, with no evidence of the large, nonparabolicity effect anticipated in the strain–split valence band. And finally, no signals were observed in the n–GaAs/(GaAl)As superlattices, despite the resonance of the quantum level splitting with the pump laser energy.

When suitably doped samples are available, we also plan studies of spin–flip nonlinearities in both InSb/(InAl)Sb and HgTe/CdTe. Calculations imply that the spin–flip signals should be observable, and will provide an excellent diagnostic of these otherwise hard–to–characterize structures.

7.6 Faraday Rotation Filters

Peter A. Wolff, Sunny Y.C. Yuen

Some years ago Romestain *et al.*[8] showed that the free carrier spin flip Raman interaction produces a large Faraday rotation of light whose energy is just below the band gap of the semiconductor. The effect was demonstrated in n–CdS, but is expected to be considerably larger in materials such as n–InSb and n–HgCdTe. Since the effect is resonant at the band gap, it can

potentially be used as the basis for an optical filter to pass near–bandgap radiation but reject other light.

To test this concept we have performed CO_2 laser, Faraday rotation experiments on n–$Hg_{0.77}CD_{0.23}Te$ crystals, whose bandgap matches the laser photon energy. Rotations of $90°$ were observed in samples 0.5 mm thick containing 2.5×10^{14} electrons/cc. When placed between crossed polarizers, such a crystal is a bandpass filter for near–gap radiation. The filter can be tuned magnetically, and might be switched with electric fields (which heat the carrier spins, thereby reducing the Faraday rotation).

These studies will next be extended to HgMnCdTe crystals where the carrier–Mn^{2+} exchange interaction provides better coupling between electron kinetic and spin temperatures.

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