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PICOSECOND SPECTROSCOPY OF HIGHLY EXCITED ELECTRONIC STATES IN NARROW GAP SEMICONDUCTORS†

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Picosecond infrared techniques have been applied to probe the kinetics of dense nonequilibrium electron-hole systems in InSb, $Hg_{1-x}Cd_xTe$, and PbTe. Specifically, time resolved transmission and reflectivity measurements have been used to examine characteristic recombination rates at excess densities of the order of $10^{19}cm^{-3}$.

The study of highly excited electronic states in narrow gap semiconductors suggests some interesting possibilities. For example, it may now be possible to employ injection from a picosecond optical source to generate a dense electron-hole system for which such conventional parameters as quasi-Fermi and plasmon energies become comparable to or larger than the single particle energy gap. In particular, with recent developments of picosecond optical sources near the 10 μm and 5 μm wavelength regions [1] it is becoming feasible to examine the time-varying dielectric properties (intraband and interband) of a dense excess electron-hole gas. We have performed first experiments in InSb, $Hg_{1-x}Cd_xTe$, and PbTe to obtain information about the characteristic decay rates at densities typically about $10^{19}cm^{-3}$ and above. At the same time measurements of some interband transition rates suggest that electron-hole correlations may make significant contributions.

In our experimental arrangement a modelocked picosecond laser at 1.06 μm ($\hbar\omega \sim 1.17$eV) served as the primary source. By employing the short pulses to operate an ultrafast infrared switch, the output from a synchronously pulsed CO_2 laser at 10.6 μm ($\hbar\omega \sim 117$meV) could be gated to generate high intensity radiation of variable duration on picosecond time scale [1].

Typically, for our purposes here, pulses of constant amplitude and 10-20 psec in duration were used and converted to 5.3 μm wavelength radiation through second harmonic generation in Te or $AgGaSe_2$ when needed. In an experiment, then, a sample of narrow-gap semiconductor was subjected to radiation from a $\sim$ 6 psec pulse of 1.17 eV photons thus leading to generation of a dense electron-hole gas near the surface. The time varying optical constant of the system was then probed by recording the transmission and reflection of controllably time delayed 10.6 μm (or 5.3 μm) pulses. Figure 1 shows results for bulk InSb at room temperature (intrinsic material). The injection of the dense excess carrier gas causes an initially rapid change (limited by experimental resolution) in reflection and transmission. This is followed by a region of approximate constancy (whose duration increases with excitation) before the onset of a time resolved decay to equilibrium. The origin of time has been chosen to coincide with the approximate beginning of the decay. Qualitatively similar data was obtained for $Hg_{1-x}Cd_xTe$ (x = .23, x = .30) and PbTe at room temperature. For comparable levels of excitation the decay was faster for $Hg_{1-x}Cd_xTe$ and slower for PbTe. In each case the intraband components ($\hbar\omega < E_{gap}$) were expected to dominate the dielectric response.

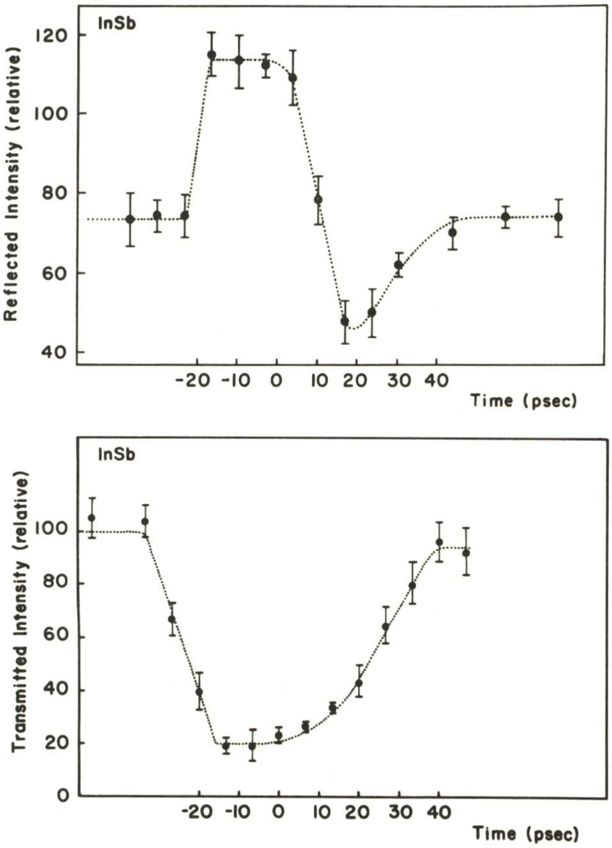

Figure 1 Time varying reflection and transmission at 10.6 μm in InSb at 300K as influenced by injection of a dense electron-hole gas

We have interpreted data such as in Figure 1 by employing a Drude-like model for a two component plasma with a $\bar{k}\cdot\bar{p}$ bandstructure in which known values for free carrier and intervalence band (not in PbTe) absorption have been extrapolated to our density regime as phenomenological damping parameters. It has been assumed that electrons and holes have thermalized with quasi-Fermi energies appropriate to lattice temperature. In the analysis we have not accounted for the influence of electron-hole scattering which may make a sizable contribution. It is noteworthy that the time resolved reflectivity shows the presence of the plasma 'dip' (not expected to be of significance in transmission due to absorption effects and the stratified nature of the medium). This permits the assignment of an approximate value for the instantaneous carrier concentration at that point. Thus for InSb we obtain an approximate decay constant of $\tau_D \simeq 30$ psec for an 'initial' excess electron density of 8×10^{18} cm^{-3}. Similarly for PbTe $\tau_D \simeq 50$ psec with an initial pair density $\Delta n = \Delta p \simeq 1 \times 10^{19} cm^{-3}$ and $\tau_D < 15$ psec for $Hg_{.7}Cd_{.3}Te$ with $\Delta n = \Delta p \simeq 1 \times 10^{19}$ cm^{-3}.

It is, of course, well known that for semiconductors such as InSb and $Hg_{1-x}Cd_xTe$ Auger recombination is a strong process, which can dominate recombination channels already at nondegenerate densities. In contrast very little experimental information is available in the highly degenerate regime [2]. Recently it has also been shown that for a multivalley "mirror band" semiconductor such as PbTe, scattering of carriers in different valleys of a band can enhance the otherwise weaker Auger rate [3]. Analysis of our data for PbTe at 300K shows that a rough agreement with a simple model can be made according to $dN/dt = -WN^{-3}$, where W is an Auger rate coefficient and equals approximately $1 \times 10^{-28} cm^{+6} sec^{-1}$. This value is consistent with earlier measurements in the nondegenerate regime, a rather surprising result considering the high degree of carrier degeneracy in our case. This is in strong contrast with our results for InSb and $Hg_{.70}Cd_{.30}Te$ which show very large deviations from extrapolations from the nondegenerate case. In a single valley model it has been pointed out by Haug [4] how degenerate statistics may significantly weaken the N^3 dependence aided further by effects of screening on the appropriate matrix elements. Further refinement of our present experimental techniques are still needed for a distinct determination of both the Auger cross-section and the concentration dependence.

An example of interband contributions is shown in Figure 2 where optical gain from a dense electron-hole gas is measured at 5.3 μm in PbTe at 20K. The temporal decay is comparable with the intraband data at room temperature.

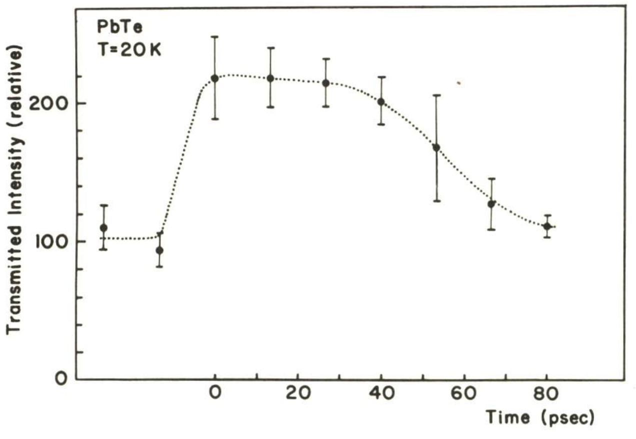

Figure 2 Time varying gain at 5.3 μm in PbTe at 20 K

On the other hand, direct calculations of maximum gain in a single particle Kane band model yield values significantly smaller than our observations. While estimates of the energy gap renormalization by exchange and correlation effects ($r_s \sim 0.1$) improve the agreement somewhat, appreciable discrepancies remain. In this connection it may be useful to note recent calculations [5] which show how Coulomb interaction in the electron-hole (excitonic) continuum can lead to enhancement in interband transition rates as observed experimentally in GaAs [6]. Extrapolation to our case may be difficult, however, because of the significantly higher injection levels in our experiments and bandstructure effects in the narrow gap semiconductors.

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