NON-LINEAR OPTICAL EFFECTS IN InSb WITH A C.W. CO LASER

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The first observations of three non-linear optical effects near the band gap in InSb at 4 K and 77 K have been made, at very low light intensities, using a c.w. CO laser with a novel attenuator system. Non-linear refraction and absorption are found to increase strongly with intensity above 30 W/cm^2 and, in addition at 4 K, the absorption decreases markedly for intensities from 1 mW/cm^2 to 30 W/cm^2 and thereafter increases. Possible origins of these effects are discussed.

We report three previously unreported non-linear optical phenomena in InSb using a c.w. CO laser. These effects, at photon energies just below the fundamental absorption edge, occur at very low beam intensities. A non-linear refraction progressively destroys the laser beam profile at intensities above 30 W/cm^2 (laser power ~ 10 mW) at 77 and 4 K. Simultaneously the absorption increases strongly. At 4 K, the absorption *decreases* for intensities between 1 mW/cm^2 and 30 W/cm^2. Essential to the experiments has been a variable laser attenuator system (Miller and Smith [1]) enabling a dynamic range of 10^5 in power to be used while maintaining a constant, gaussian beam form.

The beam from an Edinburgh Instruments PL3 CO laser, operating on any one of 60 discrete lines in the region 1670–1900 cm^{-1}, was passed through the attenuator system and focussed on to the InSb sample with spot size ($1/e^2$ intensity diameter) ranging from between 0.2 and 2 mm. The InSb sample (7.5 mm long, $N_D - N_A = 3.8 \times 10^{14}$ cm^{-3}), held in a cryostat, was antireflection coated on both of its faces to minimize Fabry-Perot fringing and to facilitate direct absolute absorption measurements, which were made possible by rotating the crystal in and out of the optical path. The *transmitted* beam was measured with pyroelectric, PbSnTe, and Ge:Cu detectors together with suitable condensing optics either to give the total power regardless of beam profile or, by mechanically scanning with a pinhole across the beam, to determine the beam's spatial profile. Both conventional and high speed (5 μs rise time) mechanical chopping systems were used, giving mark-space ratios varying from 1:1 up to 1:1000.

The non-linear refractive effect was observed at both 4 K and 77 K on all laser lines used and the typical behaviour is shown in fig. 1. This gives the diametric intensity cross-sections in the far field 18.5 cm behind the sample. The beams in absence of the sample, fig. 1d, and on transmission through the sample at low laser powers (< 5 mW) are always gaussian in cross section. However, as the incident power is increased the far field profile breaks up into a set of rings of ever-increasing radius and number. (The asymmetry in these traces may be due to slight imperfections in the incident beam.) So far it has not been possible to obtain the beam form on the exit face of the crystal so that the origin of the rings is unclear. What is certain is that

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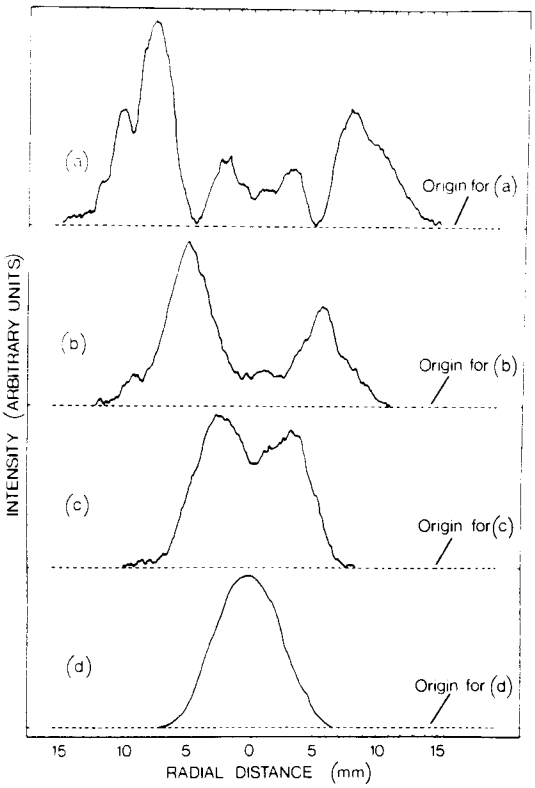

Fig. 1. Diametric beam profiles in the far field (18.5 cm) behind the samle. Incident spot 300 μm diameter, laser wavenumber 1765 cm^{-1}, temperature 77 K. (a) 41 mW incident power, (b) 25 mW, (c) 9.4 mW, (d) sample out (similar to sample in, low power).

the angular spread of the beam is increasing whether this be due to diffraction from a self-focussed spot, refraction from self-defocussing or some other effect.

We can discuss whether it is a thermal refractive effect by using the result of Akhmanov et al. [2] which gives the critical absorbed power for thermal self-focussing as well as for the onset of thermal defocussing as

$$P_T = \frac{\lambda\kappa}{dn/dT},$$

where κ is the thermal conductivity, of the order of 1 W/cm K at 77 K [3].

The temperature variation of refractive index, dn/dT, can be estimated from dispersion data [4] and thermal expansion of the band gap [5] to be $\sim 10^{-3} K^{-1}$. We have verified that the value is no greater than this by observing Fabry-Perot fringes with a similar, uncoated InSb sample as it is heated. From these data P_T is 500 mW at 5 μm, and is therefore much larger than the powers ($\sim$10 mW) at which a strong effect is observed here. The calculated value of P_T is even higher at 4 K. Thermal self-focussing/defocussing is thus unlikely on this basis. As further independent confirmation, experiments using a variable mark to space ratio chopper gave no change in the output beam form over a range of average power reaching the sample of 1:1000. In addition, no temporal effects longer than the 5 μs mechanical rise time were observed.

If we attribute the observed effects to the third order non-linear susceptibility, a value of $\chi^3(\omega: \omega, -\omega, \omega) \sim 10^{-4}$ e.s.u. is required to give the observed critical power ($\sim$10 mW) [6]. This is several orders of magnitude larger than any calculated values based on either bound electron [7] or conduction band non-parabolicity [8] contributions and the value measured by Patel, Slusher and Fleury [9] with CO_2 lasers. However, none of the calculations include *resonant*, electronic contributions due to interband transitions and the measurements [9] were made $\sim$10 μm wavelength (well away from the band gap energy). We note that strong, non-thermal, self-focussing and defocussing has been predicted [10] for the neighbourhood of an absorbing transition and has been observed at comparable laser powers in sodium vapour [11].

The detailed nature of the absorption "tail" between 1900–1700 cm^{-1} in InSb is still not interpreted with any certainty, and we therefore initiated experiments using the CO laser to measure this region, avoiding the slit resolution problems of conventional spectroscopy encountered by Roberts and Quarrington [12] and Kurnick and Powell [13]. The data in both these cases were interpreted in terms of Dumke's [14] phonon-assisted transition model, but it is not clear whether the choice of k-independent scattering potential used by Dumke is valid and the effective mass ratio required for the fit does not agree with subsequent determinations. Also, since these papers [12–14] further possible mechanisms for the existence of "tails" in III-V compounds have been advocated such as microscopic fields induced by either high impurity concentrations (for example Redfield and Afromowitz in GaAs [15]) or phonons in more ionic materials [16].

However, in attempting to carry out linear absorption measurements we have observed two distinct non-linear absorptive effects. While non-linear absorption

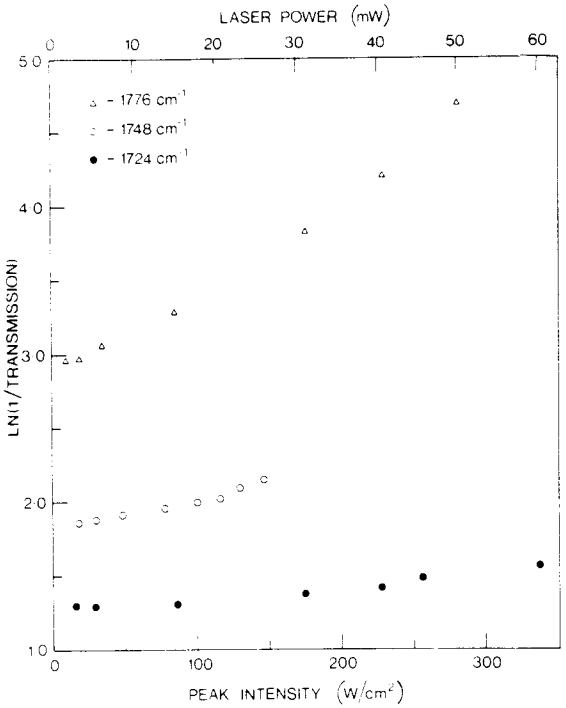

Fig. 2. Laser beam transmission of sample (7.5 mm long) at 77 K. Incident spot 210 μm diameter.

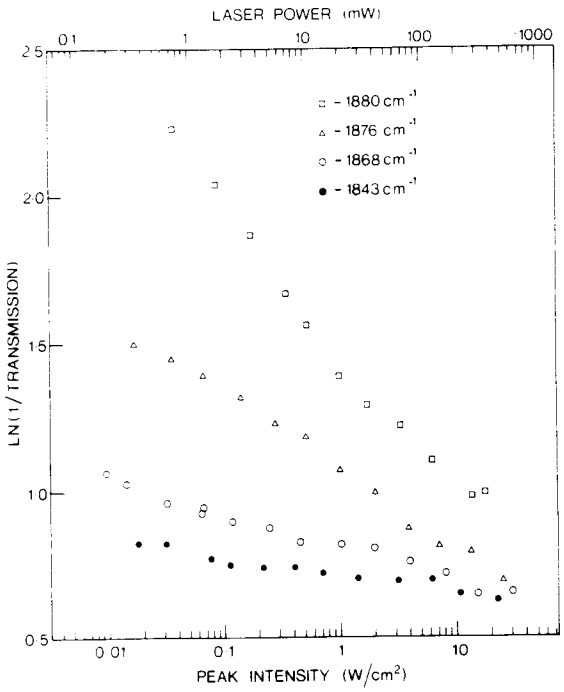

Fig. 3. Laser beam transmission of sample (7.5 mm long) at 4 K at low intensities. Incident spot 2.2 mm diameter.

ascribed to saturation has previously been observed in InSb above the band gap [17,18], the phenomena here occur below this energy.

At 77 K the absorption became non-linear above $\sim 30\,\mathrm{W/cm^2}$, increasing strongly with increasing power. Results for three typical laser lines are shown in fig. 2 and the effect is seen to be more pronounced nearer the bandgap energy (1816 $\mathrm{cm^{-1}}$ at 77 K) [19].

At 4 K while increasing non-linear absorption has also been observed above $\sim 30\,\mathrm{W/cm^2}$, the absorption behaviour near to the bandgap (1905 $\mathrm{cm^{-1}}$ at 4 K) [19] is otherwise markedly different. Linear absorption was only observed at $< 1\,\mathrm{mW/cm^2}$. Between this and 30 W/$\mathrm{cm^2}$ the absorption steadily *decreased*, as shown in the results of fig. 3 and again this effect increases dramatically nearer the bandgap giving a factor of ~ 4 increase in transmission at 1880 $\mathrm{cm^{-1}}$ between 30 mW/$\mathrm{cm^2}$ and 10 W/$\mathrm{cm^2}$.

We can estimate whether these effects could be due to local heating of the crystal. The steady state heating can be calculated on the simplifying assumption that the laser beam is a cylinder of radius w and heat is conducted through the InSb to another cylindrical surface of radius w' much larger than the beam. This gives

$$\Delta T = (P/2\pi\kappa l) \ln (w'/w).$$

Using $P \sim 10$ mW for the absorbed power, $l \sim 0.3$ cm for the length, $\kappa \sim 1$ W/cm K [3], $w = 0.01$ cm, $w' = 0.05$ cm, we obtain a temperature rise in the beam of $\Delta T \sim 10^{-2}$ K. The time taken to establish this equilibrium is given by the thermal diffusion time for a gaussian beam

$$\tau_T = \rho\, C_p\, W^2/2\kappa.$$

With density $\rho \sim 5$ g/$\mathrm{cm^3}$, $C_p \sim 0.14$ J/g K [20] we estimate $\tau_T \sim 40\,\mu$s.

If a bandgap shift due to heating of the crystal was to explain the increase in absorption at 77 K, a $\Delta T \sim 5$–10 K would be required.

This ΔT is based on calculations from the temperature dependence of the bandgap in published data [5] and confirmed by our own independent measurements. In the light of the thermal considerations of the previous paragraph such a temperature rise therefore seems very unlikely. Furthermore, experiments using the fast

chopper showed no evidence of any time constant greater than the chopper rise time (5 μs), whereas a thermal time constant $\tau_T \sim 40\,\mu$s was estimated above.

At 4 K, thermal effects are even less likely to explain the observed absorption change since not only are the onset powers of decreasing absorption very much lower (10 μW) than those discussed above, but the thermal conductivity is higher [3] and the temperature dependence of the bandgap lower [5] than at 77 K.

Thermal causes being unlikely, we must seek the explanation in terms of microscopic electronic effects: the non-linear increase of absorption may be associated, for example, with absorption by generated free carriers and the decreasing absorption at helium temperatures with a saturation effect. However, the mechanism for the linear "tail" absorption clearly requires further investigation before a plausible explanation can be advanced.

Whatever the cause of the observed phenomena, we may conclude that they will have a considerable effect on the practical operation of diode and spin-flip lasers which operate in the spectral region and the power levels where these effects are seen. Indeed anomalies in the operation of the spin-flip laser have been associated with these phenomena [21].

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