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Saturation of band-tail optical absorption in InSb

By D. A. B. Miller

Department of Physics, Heriot-Watt University, Riccarton, Edinburgh EH14 4AS, U.K.

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The optical absorption in InSb at 5 K, in the region just below the band-gap energy, is investigated as a function of the incident intensity by using a continuous wave CO laser. The linear spectrum agrees with previous measurements but saturation is seen at intensities more than about 1 W/cm^2. In the region where the linear absorption has been ascribed to acceptor-to-conduction-band transitions, the detailed variation of absorption with intensity agrees well with a saturation model developed in this paper. Nearer to the band-gap, the absorption shows some saturation at much lower intensities, and a non-saturating absorption tail is also resolved; the implications for possible absorption mechanisms are discussed.

1. Introduction

The optical absorption of the semiconductor InSb in the region about the band-gap energy has been extensively investigated, both by conventional spectroscopic methods (reviewed by Johnson 1967) and more recently with lasers (Nurmikko 1976; Lavallard *et al.* 1976). The laser studies have mostly concentrated on saturation of optical absorption above the band-gap energy, with intensities more that about 100 W/cm^2. However, the intensities used in the conventional spectroscopic measurements are likely to have been less than about 1 mW/cm^2. With new laser beam attenuation and control techniques (Miller & Smith 1978) the existence of several intensity-dependent optical phenomena has been demonstrated in the intermediate intensity range 0.001–100 W/cm^2 in cooled InSb (Miller *et al.* 1978), including nonlinear refraction at 5 K and 77 K and low intensity absorption saturation at 5 K; subsequently, on the basis of photoconductivity measurements, it was suggested that this saturation could be associated with acceptor-to-conduction-band absorption (Seiler & Hanes 1979). The nonlinear refraction is discussed elsewhere (Weaire *et al.* 1979, Miller *et al.* 1980 and 1981); in this paper the results are presented of an extended study of absorption saturation in InSb in the 'band-tail' region (i.e. just below the band-gap energy) at 5 K.

The apparatus, discussed in § 2, uses a CO laser. The low intensity linear absorption tail was measured, and a comparison with conventional measurements is given in § 3. In § 4, the results of the saturation experiments are presented and a theory of saturation of acceptor-to-conduction-band transitions is given. The comparison of experiment and theory is discussed in § 5; in particular we show that in the

region where acceptor-to-conduction-band transitions can explain the linear absorption, the agreement between experiment and theory is good, but nearer to the band-gap where the linear absorption is not understood the absorption saturation is of a different form and starts at very low intensities. As the band-gap is approached a non-saturating band-tail absorption is also resolved. The consequences for possible mechanisms of the linear and non-saturating absorption nearer the band-gap energy are discussed.

2. Experimental apparatus

The radiation source used in these experiments is an Edinburgh Instruments PL3 continuous wave CO laser, giving about 60 lines approximately equally spaced in the spectral region 5.2–6.0 μm wavelength with powers between 100 mW and 1 W. A Gaussian rather than a 'uniform' beam is used for the experiments as the Gaussian is easier to generate and control, and the necessary mathematical deconvolution for nonlinear absorption is easier to perform. To improve the beam form and enable continuously variable attenuation the beam from the laser was passed through a spatial filter–attenuator system developed for these and other related studies (Miller & Smith 1978); the resulting output is a beam of near-constant Gaussian shape and of power between about 10 μW and 100 mW. The beam is focused onto the sample with ZnSe lenses. The sample is indium-soldered to an oxygen-free copper tail in a conventional reservoir cryostat. Bulk sample heating, measured with a gold : iron-chromel thermocouple soldered to the sample, is less than 0.5 K even with 500 mW absorbed power. The sample is antireflexion-coated with a $\frac{1}{4}\lambda$ layer of ZnS on both polished faces to prevent both ordinary and nonlinear (Miller *et al.* 1979) Fabry–Perot action in the crystal. The total transmitted beam power is measured on a Laser Precision Rk 5100 Power Meter, and the sample is rotatable out of the beam to facilitate absolute absorption measurements. The material used in these experiments is n-type InSb of net carrier concentration 3.8×10^{14} cm^{-3}, tellurium-doped with no intentional p-type doping. The spacing between laser lines is about 4 cm^{-1} ($\approx$ 0.5 meV), and spectral features narrower than this cannot be resolved; there is, however, no evidence in these or previous experiments (see Johnson 1967) of such features. The use of the laser also eliminates the 'slit-width' problem encountered with conventional spectrometers when measuring an absorption coefficient changing rapidly with wavelength (where systematic errors can be introduced in absorption spectrum shape) because the spectral width of the laser line (less than about 0.003 cm^{-1}) is negligible by comparison with conventional spectrometer spectral slit-widths.

3. Linear absorption tail

The linear absorption tail in InSb at liquid helium temperatures has been investigated as a function of carrier doping by Johnson & Fan (1965). They ascribe the absorption in the region 227.6–231.3 meV to transitions between a shallow

acceptor impurity level, at an energy $E_A \approx 7.9$ meV above the valence band centre, and the conduction band; independent evidence confirms the existence of such a level and suggests it is due to a residual zinc or cadmium impurity (Putley 1959; Willardson 1967). The wavefunction of the electron associated with this impurity is (in the optically unexcited state) composed of substantially equal parts of the valence-band wavefunctions associated with k-states near band centre

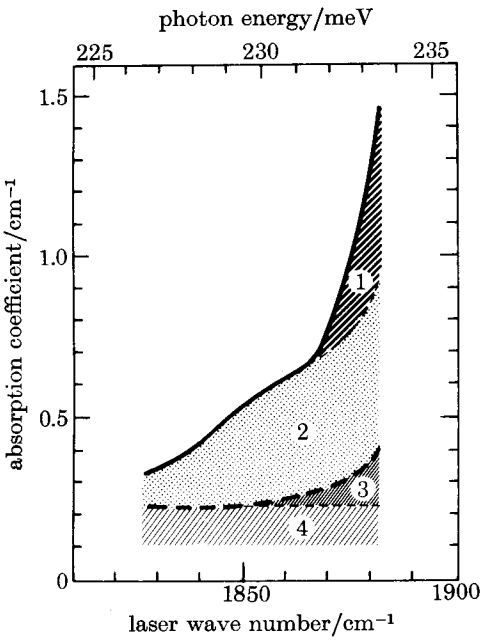

FIGURE 1. InSb absorption tail at 5 K, showing linear absorption and saturated absorption (100 W cm^{-2}) results (heavy continuous and heavy dashed lines respectively) together with speculative divisions (light dashed lines) between portions of the absorption due to differing causes. See text for an explanation of regions 1–4. The sample used was 7.5 mm long.

(Johnson & Fan 1965), the coefficients only falling off as the energy of the k-states in question are more than about 7.9 meV *below* valence band centre. This means that there is effectively no k selection rule in optical transitions from the acceptor states to the conduction band in the region below the band-gap energy. Thus a transition can take place from an acceptor level to *any* conduction band level subject only to energy conservation. Consequently, this absorption should follow the gross conduction band density of states in energy, with a threshold of about 227.6 meV, i.e. the band-gap energy E_g (nominally 235.5 meV) less the acceptor ionization energy E_a, rising thereafter with a $\Delta E^{\frac{1}{2}}$ edge ($\Delta E = \hbar\omega - E_g + E_a$). This explains the characteristic 'step' observed in the absorption tail in InSb above about 227.6 meV. This 'step' is reproduced in the results presented here (see figure 1, 'linear absorption'); in these results the absolute level of this 'step'

absorption is lower, presumably due to lower residual impurity levels in the sample.

In the results at longer wavelengths there appears to be a residual background absorption of about 0.22–0.25 cm^{-1} which does not disappear at any wavelength or intensity. This, however, corresponds to transmission of about 85% through the sample used, and the possibility that it is due to surface losses cannot be ruled out. It is presumed that it exists also in the wavelength range shown in figure 1 and is indicated as region 4 in figure 1.

At energies above about 231.3 meV in both Johnson & Fan's (1965) results and these presented here, there is a steeply rising absorption. As explained in the previous section there is no possibility in the laser measurements described here that this is an experimental artifact due to, for example, a real 'sharp' edge at $\hbar\omega = Eg$ (235.5 meV). This absorption remains to be explained. It will also be shown that this region has a distinctly different saturation behaviour from that associated with the 'step' region (below about 231.3 meV).

4. Nonlinear absorption

(a) *Results*

The dependence of absorption on intensity for four typical laser lines is shown in figure 2. As the absorption depends on the intensity it is no longer correct to use the term 'absorption coefficient' as the absorption is not obeying Beer's Law; in figure 2 the 'effective absorption coefficient' is $-l \ln (l/T)$, where l is the crystal length and T is the total fractional transmission. In the limits of low and high intensity where the transmission is again substantially independent of intensity this 'effective absorption coefficient' again corresponds to a real absorption coefficient. The peak intensity in figure 2 is the incident intensity in the centre of the Gaussian beam.

Over a range of some six orders of magnitude in intensity, the absorption on all lines is showing saturation. The lowest line, 228.7 meV, falls within the spectral region where the linear absorption can be explained by acceptor-to-conduction-band transitions, and the second lowest line, 231.3 meV, lies at the upper edge of this region; the upper two lines, 232.7 meV and 233.4 meV, lie in the region above 231.3 meV where the linear absorption rises steeply. The saturation on all four lines may look superficially similar but in fact there is a large difference between the upper two lines and the lower two lines; where significant change in absorption only occurs first in the range 0.1–1 W/cm^2 for the lower two lines, noticeable changes in the absorption of the upper two lines occur in the intensity range 0.001–0.01 W/cm^2, some two orders of magnitude lower. This distinction is clarified by the comparison with the theoretical curves also shown in figure 2. These differences will be discussed after the theory of saturation of acceptor-to-conduction-band transitions has been presented.

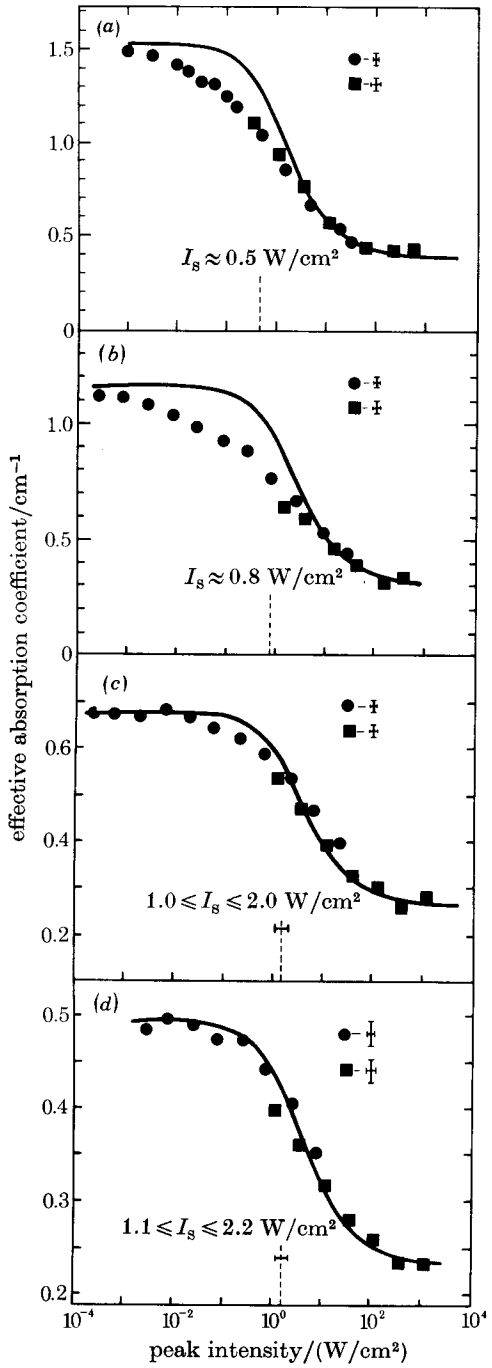

FIGURE 2. Absorption saturation results for four laser lines compared with acceptor-to-conduction-band saturation theory: (*a*) 233.4 meV; (*b*) 232.7 meV; (*c*) 231.3 meV; (*d*) 228.7 meV. Circles are taken with 1.67 mm and squares with 208 μm $1/e^2$ diameter incident beam. I_s is the saturation intensity parameter of the fitted curve. Intensities are those in the centre of the Gaussian laser beam. The same sample was used as for the results in figure 1.

(b) Theory of acceptor-to-conduction-band absorption saturation

The phenomenon of optical absorption saturation is well known in many different systems. In many situations it is the gross optical saturation resulting from the saturation of absorption in each of a large number of separate, optically non-interacting systems that is observed. Thus in the saturation of atomic absorption lines, optical transitions entirely *within* each separate localized atom are saturated; there are no optical transitions *between* the separate atoms. For transitions between semiconductor bands, such as have recently been investigated in germanium (see James & Smith 1980, and literature cited therein), a similar formalism can be used for the extended (i.e. non-localized) band states, though for entirely different reasons; the usual selection rule of 'vertical' (i.e. k-conserving) optical transitions allows each value of k to be considered as if it were a separate system or 'atom', there being no optical transitions *between* the different k-values, the only optical transitions being *within* the states of identical k-values in different bands. However, the situation with acceptor-to-conduction-band transitions is different to both of these extreme cases; each acceptor may interact optically with many conduction-band states, and each electron in a conduction-band state may relax to any acceptor state, but it is relatively simple to derive a saturation model for this case through a rate equation analysis, and the final result is similar to standard saturation relations.

As electrons are excited from acceptor states into the conduction-band k-states, they will relax rapidly towards a thermal distribution. This phenomenon has already been reported for InSb with interband transitions (Lavallard *et al.* 1977). At these low temperatures and carrier concentrations the electrons will therefore collect very close to the bottom of the conduction band. Provided we avoid this region we may assume therefore that the occupation probability of conduction-band states is negligible. Therefore the total transition rate from an acceptor to the conduction band can be described by

$$W_{\mathrm{a.c.}} \simeq (I/\hbar\omega)\, P_{\mathrm{a}}\, \sigma_{\mathrm{c}}, \tag{1}$$

where σ_{c} is the total linear absorption cross-section for absorption from an acceptor to the conduction band, P_{a} is the occupation probability of the acceptor state, I is the radiation intensity and $\hbar\omega$ is the photon energy. (This is both the gross and the net induced radiactive transition rate as the upper-state population is assumed zero and hence the normal stimulated emission term is also zero).

We assume a simple phenomenological decay constant τ_{r} for relaxation of the excited electrons back down to the acceptor states and that the net downward transition rate is also proportional to the probability, $1 - P_{\mathrm{a}}$, of finding the particular acceptor state empty. The problem is now readily solved for the occupation probability P_{a} in the usual manner for the steady state by balancing the rates of excitation and decay.

If there are N_{a} acceptors per unit volume altogether, effectively all of which will

initially be occupied at 5 K, then the linear absorption coefficient α_l of acceptor-to-conduction-band absorption will be

$$\alpha_l = N_a \sigma_c,$$

and the nonlinear absorption coefficient $\alpha_{n.l.}$ ($\equiv -I^{-1}\mathrm{d}I/\mathrm{d}z$) $= \alpha_l P_a$ is finally

$$\alpha_{n.l.} = \alpha_l/(1+I/I_s), \tag{2}$$

where

$$I_s = \hbar\omega/\sigma_c\tau_r = \hbar\omega N_a/\alpha_l\tau_r. \tag{3}$$

Thus we expect acceptor-to-conduction-band absorption to saturate like homogeneous saturation of a two-level system.

To compare this theory with experiment the variation of the nonlinear absorption coefficient through the crystal (due to the variation of I by nonlinear absorption), and across the initially Gaussian profile, must be included. The profile of the beam will alter as it propagates through the crystal because some parts of the profile will be absorbed more strongly than others. In principal also the modified profile will diffract inside the crystal, but in practice we shall neglect this; the diffraction length of a beam half the diameter of the smallest beam used experimentally (208 μm) is only just of the order of the crystal length (7.5 mm). Neglect of diffraction means that each part of the beam can be treated separately, with no interaction with other parts of the profile, and the results for each initial intensity in the profile integrated to give the total transmission of the whole beam.

The nonlinear absorption equation for one part of the beam, expressed in dimensionless form and including a background non-saturating linear absorption, α_b, is

$$\mathrm{d}\psi/\mathrm{d}\zeta = -[1/(1+\psi), +\gamma]\psi, \tag{4}$$

where $\psi = I/I_s$, $\zeta = \alpha_l z$ and $\gamma = \alpha_b - \alpha_l$. The solution of this equation is given by

$$\frac{\psi_l}{\psi_0}\left[\frac{\eta+\psi_l}{\eta+\psi_0}\right]^{1/\gamma} = \mathrm{e}^{-(\gamma+1)\zeta_l},$$

where $\eta = (1+\gamma)/\gamma$, $\zeta_l = \alpha_l l$ (l is the crystal length), and ψ_0 and ψ_l are the incident and transmitted (dimensionless) intensities respectively.

Equation (5) can be solved numerically to give ψ_l in terms of ψ_0, γ and ζ_l, i.e. $\psi_l = f(\psi_0, \gamma, \zeta_l)$. For a Gaussian beam

$$\psi_0 = \psi_p \mathrm{e}^{-\rho_2},$$

where ψ_p is the (dimensionless) intensity in the beam centre, $\rho = r/w$, r being the radial distance from the beam centre, and w is the $1/e$ intensity beam radius. We obtain the ratio of transmitted to incident power, T, by integrating over the entire beam:

$$T = \frac{1}{\psi_p}\int_0^\infty f(\psi_p \mathrm{e}^{-\rho^2}, \gamma, \zeta_l)\, 2\rho\, \mathrm{d}\rho. \tag{6}$$

To fit this theory to experiment, the parameters γ and ζ_l are calculated from the minimum and maximum transmissions, a curve of T against ψ_p is plotted on a logarithmic intensity scale, and the best value of I_s is deduced by adjusting the position of the curve on the intensity axis for best fit of curve and experimental points. Note that the curves are determined by only three parameters; with the minimum and maximum transmissions chosen the shape of the curve is fixed and only the value of I_s remains adjustable.

The experimental results on the saturation of band-tail absorption for the four laser lines are compared in figure 2 with theoretical curves from the above model. For the two lower-energy lines (228.7 meV and 231.3 meV) the theoretical curves are best fits, whereas for the higher-energy lines (232.7 and 233.4 meV) the theoretical curves are approximately fitted only to the higher intensity data (more than 10 W/cm^2) to show the breakdown of the form of the saturation at low intensities on these lines.

5. Discussion

(*a*) *Acceptor-to-conduction-band absorption saturation*

For the two lines (228.7 and 231.3 meV) that lie within the region where the linear absorption is explained by acceptor-to-conduction-band transitions, the agreement of saturation theory and experiment is good, with fitted saturation intensities of about 1–2 W/cm^2. Predicting the absolute value of the saturation intensity, I_s, is difficult because neither the residual acceptor concentration, N_a, nor the recombination time τ_r is known with any precision. However, N_a and τ_r are linked through equation (3) and we can estimate N_a from the strength of the linear absorption from acceptors to the conduction band. Comparing the height of the acceptor absorption 'step' in the results presented here (corrected for background absorption) with the data of Johnson & Fan (1965) and scaling proportionately from their quoted acceptor concentration gives $N_a \approx 10^{14}$ cm^{-3}, whereas calculating N_a from the same (corrected) absorption strength by using Dumke's (1963) model gives $N_a \approx 10^{13}$ cm^{-3}. The discrepancy between these estimates remains to be explained and we note that no attempt is made by Johnson & Fan (1965) to calculate the absolute absorption strength. Using equation (3) with a saturation intensity of about 1.5 W/cm^2 implies that for $N_a \approx 10^{14}$ cm^{-3}, $\tau_r \approx$ 8 μs, while for $N_a \approx 10^{13}$ cm^{-3}, $\tau_r \approx$ 800 ns. Interband recombination times are known to be of the order of hundreds of nanoseconds in InSb under these conditions (Fossum & Ancker-Johnston 1973), suggesting $N_a \approx 10^{13}$ cm^{-3} rather than 10^{14} cm^{-3}. On the basis of this discussion, however, we can at least conclude that the measured saturation intensities are in order-of-magnitude agreement with expected values for the 228.7 and 231.3 meV lines.

(*b*) *Other components of band-tail absorption*

The experimental data, in addition to confirming the acceptor saturation model, also reveal other aspects of the absorption edge of InSb at 5 K and suggest that this edge is composed of a variety of parts, illustrated in figure 1 (over a background absorption, region 4, assumed uniform).

(*a*) The high intensity 'saturated' absorptions on different lines do not all tend to the same level; there is a distinct rise as the band-gap is approached (region 3, figure 1).

There is independent empirical evidence for the existence of this non-saturating absorption. (i) Photoconductivity results (Hanes & Seiler 1979) in the region just below the bandgap in InSb show the photoconductive signal continues to rise at intensities above about 50 W/cm^2 and is larger for lines nearer the band-gap energy. (ii) Measurements (Miller *et al.* 1979) at intensities of about 100–200 W/cm^2 at a laser line nearer the band-gap energy (235.0 meV) indicate a substantially linear absorption over this high intensity range of the order of 10 cm^{-1}. It is reasonable to assume that this is the continuation of the trend of rapidly rising non-saturable absorption.

An absorption such as this which does not appear to saturate, or at least saturates only at some higher intensity, can be explained if it couples large numbers of states (or states with short relaxation times, or both). Thus one possible mechanism for this absorption is indirect interband absorption, assisted by some scattering event (e.g. carrier–carrier or carrier–phonon scattering); because the k-selection rule, and the requirement that the initial and final states be separated by the photon energy, are now both removed by the scattering process, large numbers of states in valence and conduction band are now coupled optically, albeit through a weak process. Such an absorption can only be saturated if this large number of states is populated, requiring correspondingly high intensities. Carrier–phonon scattering is unlikely to explain the detailed shape of this non-saturating absorption, however, because optical phonon scattering could not explain the sharp resonance as the band-gap energy is approached (the optical phonon energy being about 24 meV), and single acoustic phonon scattering would be too resonant owing to the requirement of energy and momentum conservation (restricting such scattering events to within less than 10 μeV of the band edge). However, carrier–carrier scattering remains a strong possibility.

Theoretically (Johnson & Fan 1965) the acceptor-to-conduction-band absorption (region 2, figure 1) should continue to rise slowly above 231.3 meV and on the assumption that it is a separate mechanism from the non-saturating absorption (region 3) the light dashed line between regions 1 and 2 has been sketched by adding the theoretical rise in acceptor absorption to the measured non-saturating absorption (region 3).

(*b*) A further absorption component (region 1, figure 1) is required to explain the linear absorption spectrum. Also, in this spectral region above 231.3 meV, the

absorption saturation is qualitatively different and begins at very much lower intensities as shown by the comparison (figure 2) at 232.7 and 233.4 meV. (There is also some indication of the start of this behaviour in the 231.3 meV line at 0.1–1 W/cm^2).

While a full explanation of this absorption is beyond the scope of this paper, we can draw some conclusions about its cause from the saturation behaviour. Clearly some part of this absorption saturates at very low intensities, with significant change in absorption being seen at about 0.01 W/cm^2 at 232.7 and 233.4 meV (compared with about 1 W/cm^2 at 231.3 meV and 228.7 meV).

Low saturation intensities imply that the number of states involved in the saturation is small or the recombination time is large, or both. It is therefore unlikely that this absorption is due to ordinary interband transitions because (i) any steady-state interband saturation must involve at least the number of states within kT, which even for the very bottom of the conduction band in InSb is about 10^{14} cm^{-3} at 5 K; (ii) the band-to-band recombination time is known to be of the order of hundreds of nanoseconds.

These numbers would indicate a saturation intensity of the same size or larger than that for acceptor-to-conduction-band absorption. One possible explanation for this absorption is transitions from a density-of-states tail on the valence bands to the conduction band, as then very small numbers of states are involved. For example, by using the Thomas–Fermi model given by Kane (1963) for density-of-states tails in impure semiconductors with an impurity concentration of about 4×10^{14} cm^{-3}, for energies greater than about 1 meV above the valence-band-edge there are altogether about 10^{13} cm^{-3} tail states while above about 1.5 meV there are only about 5×10^{10} cm^{-3}. However, to explain the linear absorption, the absorption cross sections of these states would need to be correspondingly higher. Hanes & Seiler (1980) have also postulated the existence of an acceptor level 3 meV above the valence band on the basis of photoconductivity measurements (although the intensities used in their experiments were much greater than 1 W/cm^2). A low concentration of such acceptors with correspondingly higher cross sections could also explain the absorption in region 1 and its saturation. While exciton resonances are not observed in InSb without a magnetic field, presumably because the electron–hole interaction is readily screened even for free carrier densities of about 10^{14} cm^{-3} (the calculated exciton orbit radius is about 70 nm), the possibility that this absorption is linked to some remanent coulomb interaction between electron and hole also cannot be ruled out, although no model for such an absorption and its saturation behaviour appears to exist at present.

6. Conclusions

In conclusion, we have demonstrated that the band-tail optical absorption in InSb at 5 K can be saturated with modest laser intensities, that this saturation can be explained by a simple model for the spectral region where linear absorption is

already explained by acceptor-to-conduction-band transitions and that the saturation behaviour enables us to distinguish two further regions of absorption (one of which, the 'saturated absorption', cannot be resolved in linear absorption experiments), and to make general deductions about their causes.

The different saturation behaviour of absorptions seen here in InSb suggests that absorption saturation may be a useful technique for characterizing the different absorptions and their causes near the band-gap of semi-conductors.

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