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High-resolution two-photon spectroscopy in InSb at milliwatt cw powers in a magnetic field

D. G. Seiler* and M. W. Goodwin
Department of Physics, North Texas State University, Denton, Texas 76203

M. H. Weiler
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
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Resonant structure in the magnetophotoconductivity of n-InSb is shown to arise from two-photon absorption of cw CO_2 laser radiation. This is the first time that two-photon experiments in solids have been carried out with only cw lasers.

Weak magneto-optical transitions in high-purity n-InSb induced by a CO_2 laser have been recently observed by photoconductivity measurements for $\vec{e} \| \vec{B}$ in the Voigt configuration for magnetic fields up to 1.2 T.[1-3] This resonant structure was first attributed to multiple LO phonon emission from high-energy photoexcited electrons created by intraband absorption.[1,2] Later, it was proposed that the structure instead originates from midgap deep defect levels that are known to be present in InSb.[3] Two-photon resonant magneto-absorption was not suspected because of the use of cw lasers and the very low intensities (≈ 1 W/cm^2) where the structure can first be observed. In this paper we present new experimental results on this resonant structure for $\vec{e} \perp \vec{B}$ and for fields up to 2 T. The simultaneous use of two CO_2 lasers has enabled us to identify unambiguously the physical origin of these resonant transitions as two-photon absorption rather than either of the two previous interpretations. This is a surprising and important result since two-photon effects (in any material) have only been previously observable with the help of at least one high-power pulsed laser which produces the high photon fluxes that were previously thought to be necessary. The use of cw lasers for two-photon absorption opens up new opportunities in spectroscopy by allowing high-resolution studies of new transitions with selection rules and exciton effects which are different from one-photon transitions.

The evidence for the two-photon-absorption interpretation which we present in this paper consists of (1) the change in conductivity induced by two cw CO_2 lasers simultaneously incident on the sample, (2) the amplitude of the structure which varies as the square of the intensity, and (3) the good agreement between theoretical and experimental two-photon transition energies. Our new experimental results cannot be explained by the multiple-phonon or deep-level models since neither model is consistent with the two-laser results or the intensity dependence of the amplitudes.

In our experiments, a constant dc electrical current is applied to the sample (of concentration 9×10^{13} cm^{-3}) while a small ac magnetic field modulates the sample conductivity at a frequency of 43 Hz. The photoconductive signal produced by the chopped laser (~ 20-μsec pulse width and 1700-Hz repetition rate) is fed into a sampling oscilloscope, the output of which is fed into a lock-in detector with response proportional to the second derivative of the photoconductive signal. All experiments were done in the Voigt configuration with either $\vec{e} \perp \vec{B}$ or $\vec{e} \| \vec{B}$ and the current parallel to $\vec{B}$ with $\vec{B}$ parallel to a $\langle 110 \rangle$ crystallographic direction.

The crucial experiment that clearly shows that the observed resonant structure in the photoconductivity is due to two-photon processes is illustrated in Fig. 1. The top curve shows a photoconductive trace ob-

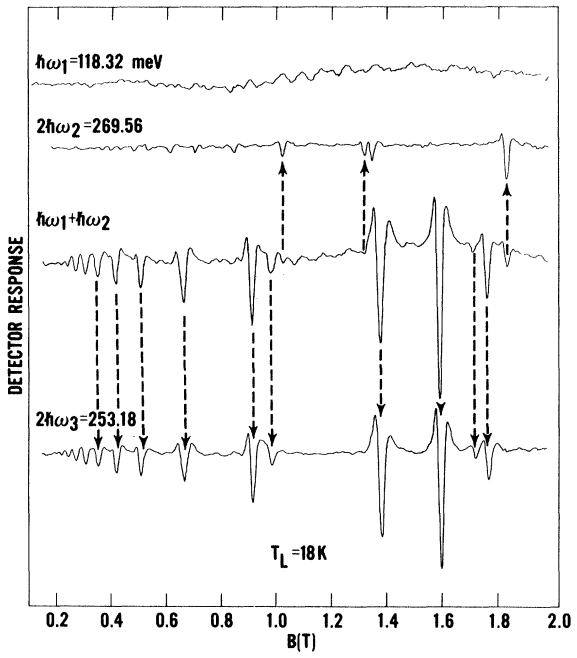

FIG. 1. Photoconductive spectra with $\vec{e}\perp\vec{B}$ at $T_L = 18$ K using various combinations of two-photon energies: $2\hbar\omega_1 = 236.64$ meV, $2\hbar\omega_2 = 269.56$ meV, $\hbar\omega_1 + \hbar\omega_2 = 253.10$ meV, and $2\hbar\omega_3 = 253.18$ meV. The spectrum with $\hbar\omega_1 + \hbar\omega_2$ was obtained with two separate laser beams simultaneously incident upon the sample.

tained from a single 10.48-μm ($\hbar\omega_1 = 118.32$ meV) laser operating with a large peak incident power of 1.8 W. The sharp resonant structure is absent, and only one-LO phonon-assisted cyclotron harmonic transitions within the conduction band can be seen.[4] The next trace shows the resonant structure obtained with a single 9.20-μm line ($\hbar\omega_2 = 134.78$ meV) at a lower power of 0.2 W from another laser. The two-beam experimental results are shown in the next trace where completely different structure is seen, with only traces of the structure due to the weak 9.20-μm laser. Clearly this new structure arises only because of the presence of the two photons $\hbar\omega_1$ and $\hbar\omega_2$. As a final check, if two-photon processes are the origin of this new structure, the positions are determined by $\hbar\omega_1 + \hbar\omega_2$. Consequently, the same structure should be observed with only one laser, but at a different photon energy $\hbar\omega_3$ such that $2\hbar\omega_3 = \hbar\omega_1 + \hbar\omega_2$; the last trace shows that this is in fact the case using only one laser operating at 9.795 μm ($2\hbar\omega_3 = 253.18$ meV).

The only other possible interpretation of these data, aside from two-photon absorption, could be absorption of second-harmonic intensity produced when the incident radiation is polarized along a ⟨110⟩ crystal axis. However, we have also observed these same resonant effects for light polarized along a ⟨100⟩ axis. Consequently, we conclude that second-harmonic effects are negligible and that we are observing pure two-photon resonant transitions.

The amplitude of the resonant structure, which is directly related to the number of photoexcited carriers, was found to vary approximately as the square of the intensity. This is additional confirmation of the involvement of two-photon absorption. The number of photoexcited carriers n created by two-photon absorption is given by

$$\frac{dn}{dt} = A_2 I^2 - r_2 n(n + n_0) \quad , \tag{1}$$

where A_2 is the coefficient of two-photon absorption,[5] r_2 is the coefficient of quadratic or direct recombination,[6] I is the light intensity incident on the sample, and n_0 is the electron density when $I = 0$. For the small intensities of our experiments where $n \ll n_0$, we then have for steady state $n \simeq (A_2/r_2 n_0) I^2$, which explains the observed intensity dependence of the amplitude.

For the purpose of obtaining a value for A_2, we repeated our experiments using a boxcar integrator with no field modulation. From the photoconductivity voltage at resonance we estimate that $n \approx 1 \times 10^{13}$ cm^{-3} at a peak intensity of ~ 130 W/cm^2 for a peak observed at $\lambda = 9.59$ μm at $B \approx 1.2$ T. Averaging over a Gaussian intensity distribution and using a value of $r_2 \approx 1 \times 10^{-9}$ cm^3/sec,[6] and $n_0 \simeq 9 \times 10^{13}$ cm^{-3}, gives $A = 0.7$ cm sec/erg^2 in reasonable agreement with the experimental results of 1.6 cm sec/erg^2 measured by Nguyen *et al.*[5] for the same line observed in absorption. This value is also consistent with their theoretical estimate. A more detailed analysis of the intensity will be published later.

There have only been a few previous observations of multiphoton absorption in InSb in a magnetic field, first by Button *et al.*[7] This work was extended by Weiler *et al.*,[8] who interpreted the data using a tunneling theory of multiphoton absorption[9] which, like the zero-magnetic-field theory of Keldysh,[10] was later shown to be incorrect for two- (or even-) photon absorption.[11,12] Manlief and Pakik[13] explained most of their experimental results using the selection rules from perturbation theory, which were later also given by Zawadzki and Wlasak[14] with the absorption for polarizations $\vec{e}\parallel\vec{B}$ also observed and explained by Favrot *et al.*[15] Nguyen and co-workers[5] also explained their observation of two-photon absorption for $\vec{e}\perp\vec{B}$ with transition strengths calculated from perturbation theory using nonparabolic wave functions. The most recent study of two-photon absorption in InSb, in zero magnetic field, is that of Pidgeon and co-workers,[16] who give references to other work, and show that a perturbation-theory treatment, taking into account the exact nonparabolic wave functions and neglecting exciton effects, gave good agreement with their experimental results. Their conclusions

should also be valid for experiments in a magnetic field.

Comparison of our data shown in Fig. 1 with the two-photon data of Ref. 13 (Fig. 1) using a transversely excited atmosphere (TEA) CO_2 laser and of Ref. 5 (Fig. 1) using a q-switched CO_2 laser clearly illustrates the high resolution of our measurements where structure is seen at fields as low as ~ 2 kG. Also, an apparent doublet structure is resolved in some lines which has not previously been observed. We have observed two-photon structure using a single laser with peak incident powers as low as 14 mW in a beam of 1.5-mm $1/e^2$ intensity diameter.

Further confirmation of the identification of the resonant structure with two-photon absorption is provided in Fig. 2, where the resonance positions, from experiments with a single laser, are compared with a theoretical calculation. The theoretical transition energies were calculated using a refinement[17] of the Pidgeon and Brown 8×8 model[18] with parameters adjusted to give excellent agreement with interband and intra-conduction-band magneto-optical experiments. A value of $E_g = 236.6$ meV gave the best overall fit, with the other band parameters as given in Ref. 17. In Ref. 17 the observed one-photon interband transition peaks were identified with the exciton ground state associated with each transition by reducing the calculated interband transition energy by an approximate exciton binding energy.[19,20] For comparison with the present experiments we have used a numerical approximation[21] to the calculated exciton binding energy which is valid for the range of magnetic field values in Fig. 2.

The theoretical curves in Fig. 2 were calculated using the two-photon perturbation theory selection rule $\Delta n = 0, \pm 2$.[5,14,15] The positions are quite different from those with the single-photon selection rule $\Delta n = \pm 1$, differing essentially by a hole or electron cyclotron resonance energy. The high resolution of the present data provides an opportunity for identification of individual two-photon transitions, as opposed to unresolved groups of transitions. The fact that new transitions are observed is a general characteristic of two-photon spectroscopy, and provides a further test and opportunity for refinement of the theoretical model.

It is clear from an examination of Figs. 1 and 2 that the experimental resolution of 0.2 meV is sufficient to resolve not only the exciton ground-state peaks, but also excited-state peaks, associated with the different interband transitions. In Fig. 2 only the exciton ground-state transitions are plotted. Although the exciton correction is quite crude, we are satisfied with the good overall fit with the data. In future work we will attempt a better identification of the observed lines using an improved exciton correction, taking into account the possibility of excited exciton states. This will also allow a more accurate determination of the band parameters from this complex set of two-photon data.

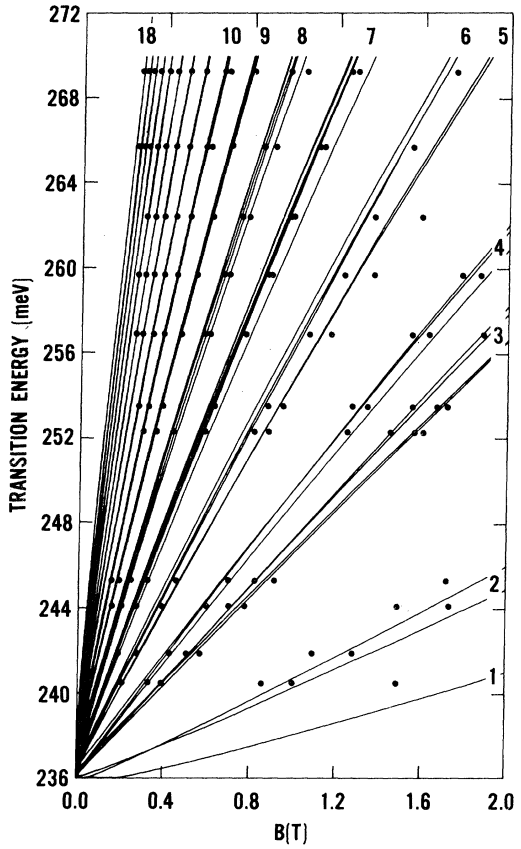

FIG. 2. Transition energies vs magnetic field. The dots represent the experimental data while the curves are the transition energies calculated for the strongest allowed two-photon transitions $\Delta n = 0, \pm 2$. We give below, in the notation of Ref. 17, the strongest transitions corresponding to each group of curves: 1: $a^-(2) \rightarrow a^c(0)$; 2: $a^+(0) \rightarrow a^c(0)$, $b^-(2) \rightarrow b^c(0)$; 3: $a^+(-1), a^-(3) \rightarrow a^c(1)$, $b^+(0) \rightarrow b^c(0)$; 4: $b^+(-1), b^-(1), b^-(3) \rightarrow b^c(1)$; 5: $a^+(2) \rightarrow a^c(0)$; 6: $a^+(1) \rightarrow a^c(1)$, $a^-(4) \rightarrow a^c(2)$, $b^-(2) \rightarrow b^c(2)$; 7: $a^-(5) \rightarrow a^c(3)$, $b^-(1), b^-(3) \rightarrow b^c(3)$, $b^+(1) \rightarrow b^c(1)$, $b^+(0) \rightarrow b^c(2)$; 8: $a^-(6) \rightarrow a^c(4)$, $b^-(4) \rightarrow b^c(4)$, $a^+(2) \rightarrow a^c(2)$, $a^+(3) \rightarrow a^c(1)$; 9: $a^-(7) \rightarrow a^c(5)$, $b^-(3), b^-(5) \rightarrow b^c(5)$, $b^+(2) \rightarrow b^c(2)$, $b^+(1) \rightarrow b^c(3)$; 10: $a^-(8) \rightarrow a^c(6)$, $b^-(6) \rightarrow a^c(6)$; $a^+(3) \rightarrow a^c(3)$, $a^+(4) \rightarrow a^c(2)$; etc.

In summary, we have carried out the first two-photon experiments in solids using only cw lasers. A new dimension for two-photon spectroscopy has now been created with the distinct advantages that use of cw lasers has to offer: (1) better long- and short-term amplitude stability than pulsed lasers; (2) enhancement of signal-to-noise with the use of modulation and lock-in amplifier techniques. Other semiconductors with different band gaps will undoubtedly be studied with the appropriate cw lasers in the near future.

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*Present address: Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Mass. 02139.

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