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FAR-INFRARED TWO-PHOTON INTRABAND TRANSITIONS IN n-InSb

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Two-photon cyclotron resonance and two-photon shallow donor transitions have been excited by a high power pulsed D_2O laser emitting λ = 119 µm and 66 µm laser lines at the same time. Transitions involving the absorption of two 119 µm photons or simultaneously one 119 µm and one 66 µm photon were observed. Two-photon selection rules are discussed by a rigorous treatment of the symmetry of the free electron Hamiltonian.

I. Introduction

The development of high power far-infrared (FIR) molecular lasers initiated a growing number of nonlinear optical investigations in the long wavelength infrared spectral range including the saturation of cyclotron resonance and shallow impurity transitions in semiconductors [1-5] and second harmonic generation [6]. FIR two-photon transitions between 1s-2s shallow donor levels and two-photon cyclotron resonance were first observed in n-GaAs with the highly sensitive method of magneto-photoconductivity [7] In this paper we report first experimental results concerning two-photon intraband absorption in n-InSb and their interpretation by means of two-photon selection rules. Evidence for two-photon transitions stems from the good agreement of observed peaks in magneto-photoconductivity with calculated energy separations and from the nonlinear power dependence of the signals.

II. Experimental Technique

The measurements were carried out at liquid helium temperature in a superconducting magnet. The sample was a 0.5 mm thick high purity n-InSb crystal of $N_D - N_A = 9 \cdot 10^{13}$ cm^{-3} effective donor concentration. It was mounted in an integrating cavity with the [100] crystallographic direction parallel to the magnetic field. The beam of a TEA-CO_2 laser pumped D_2O laser emitting radiation of 66 µm and 119 µm wavelengths at the same time was transmitted through a metallic wave guide which was terminated by a cone to concentrate the radiation on the sample. A small fraction of the power decoupled by a beam splitter was detected by a n-GaAs photoconductor of 2 ns response time to monitor the peak power. The GaAs detector was calibrated by a pyroelectric energy meter which was assumed to be linear in the power range of the measurement. Photoconductivity was measured by irradiating the power of both laser lines on the samples or by using the 119 µm line only. In the latter case radiation of 66 µm was eliminated by a cold KCl single crystal filter. The peak power of the total emission was about 10 kW and that of the 119 µm line after filtering was approximately 2 kW The power was varied by an aperture stop of variable diameter reducing the power of both laser lines in the same way.

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III. Results

In Fig. 1 the measured photoconductive signal excited by both wavelengths is plotted as a function of the magnetic field strength B for various irradiation peak powers. In order to identify the observed peaks the magnetic field dependence of the two lowest Landau levels for both spin orientations calculated with the Trebin-Rossler-Ranvaud model [8] and shallow donor transition energies as far as known from literature [9] are included in Fig. 1. Possible one- and two-photon transitions coincide with observed structures in photoconductivity and are indicated by arrows. The cyclotron resonances $0^+ \rightarrow 1^+$ and $0^- \rightarrow 1^-$ - corresponding to $(2^-2^-00) \rightarrow (3^+3^+11)$ and $(1^-1^-11) \rightarrow (2^+2^+20)$, respectively, in the full notation of ref. [8] - and the impurity transitions $(000)^+ \rightarrow (110)^+$ give rise to strong one-photon signals around B = 1 4 T and 2.6 T for λ = 119 µm and 66 µm, respectively The individual lines are not resolved due to the rapid saturation of one-photon signals particularly at low magnetic field strength for λ = 119 µm and because of the large thickness of the sample, which was chosen to facilitate the detection of weak structures in photoconductivity Above about B = 3 T no one-photon transition is expected to occur In this magnetic field range the signal depends superlinearly on the exciting power Peaks 1 and 2 (Fig. 1a) agree well with the two-photon cyclotron resonance transitions without spin flip $0^+ \rightarrow 1^+$ and $0^- \rightarrow 1^-$, respectively, involving two 119 µm photons An impurity transition (119 µm and 66 µm) may also contribute to peak 2 The weak structure denoted by 3 in Fig 1a just corresponds to the magnetic field strength of two-photon cyclotron resonance $0^+ \rightarrow 1^-$ with spin flip generated by simultaneously absorbing one 119 µm and one 66 µm quantum

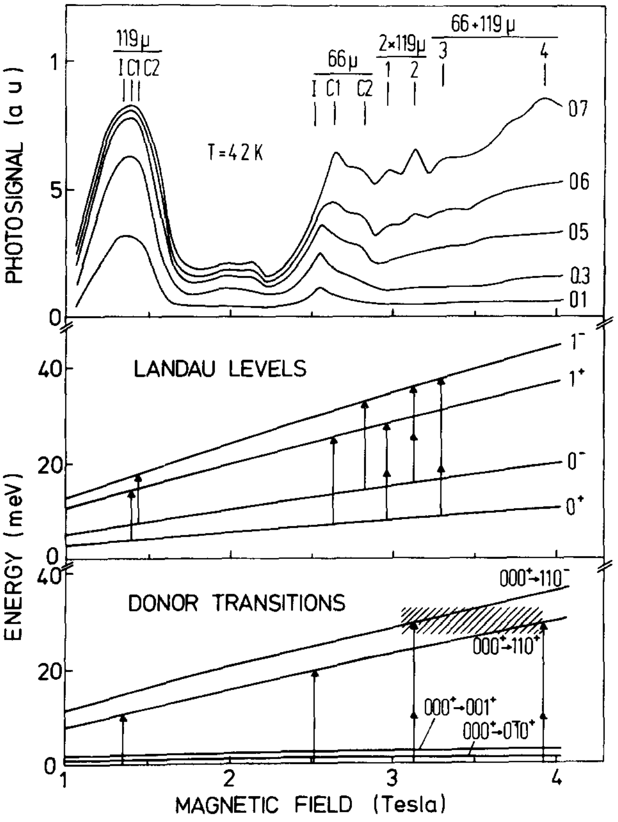


Fig 1 (a) Magneto-photoconductivity of n-InSb excited by the λ = 119 µm and 66 µm lines of a D_2O laser. Numbers on the right side denote laser power in relative units, 1 corresponds to about 10 kW (b) Landau levels after [8] (c) Shallow donor transition energies after [9] Single and double arrows in (b) and (c) indicate possible one- and two-photon transitions, respectively

In the range of B between 3 3 T and 4 T (shaded in Fig 1c) the photoconductive signal must be generated by two different photons (119 µm and 66 µm) being due to transitions from the donor energy level series $(00\beta)^+$ and $(0\bar{1}\beta)^+$ below the lowest Landau level to final states in the series $(11\beta')^-$ and $(11\beta')^+$ The involved donor bound state quantum number β, β' cannot be recognized because of the low resolution of the measurement resulting from the large thickness of the sample

In Fig. 2 the photosignal of the λ = 119 µm line alone is shown for various peak powers between B = 2.5 T and 3.5 T In this range of magnetic field strength no resonant one-photon transitions are possible with radiation of this wavelength The lines 1 and 2 are again identified as two-photon cyclotron resonances $0^+ \rightarrow 1^+$ and $0^- \rightarrow 1^-$, respectively. The peaks 5 and 6 may be attributed to two-photon absorption of shallow donors These transitions are hidden in Fig 1 by the strong 66 µm one-photon excitation

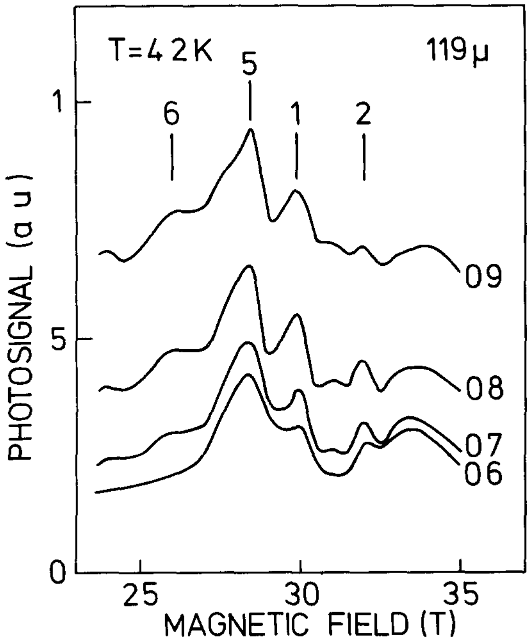


Fig 2. Magneto-photoconductivity measured for λ = 119 μm Numbers on the right side denote irradiation power in relative units, 1 corresponds to about 2 kW

IV Two-Photon Selection Rules

Two-photon transitions require intermediate states being connected by electric dipole matrix elements with the initial and final states of the transition Thus selection rules for two-photon transitions between Landau levels (free states) or between impurity levels attached to Landau levels (bound states) can be obtained from the corresponding one-photon selection rules. Dipole selection rules for transitions between free states have been formulated rigorously on group-theoretical grounds [8]. The dipole operator can be represented as the derivative of the Hamiltonian with respect to the Landau operators a^+ (cyclotron resonance active polarization e_+), a (e_-) or the momentum component ζ parallel to the magnetic field (e_3). Therefore, as the Hamiltonian contains parts with axial, cubic or tetrahedral symmetry, dipole transitions become allowed with the weight M_i, i = 0,.. ,5 of these terms. M_o ($e_\pm$) type transitions follow from axial symmetry and correspond to a change in the Landau quantum number by ±1 M_2 can be classified as warping induced and M_4 as inversion asymmetry induced transitions While M_o, M_2 and M_4 denote transitions at $\zeta = 0$, those of type M_1, M_3 and M_5, respectively, refer to $\zeta \neq 0$ transitions. Fig. 3 shows possible two-photon transitions for cyclotron and combined resonances It turns out that combined two-photon resonance is possible only with two quanta of $e_\pm$ polarization, while two-photon cyclotron resonance is possible only, if one photon is e_3 polarized. The probability of type M_2 .M_5 transitions owing to their cubic or tetrahedral symmetry should depend on the orientation of the magnetic field with respect to the crystallographic axes [10] This could be shown in a properly designed experiment

Donor states in small gap semiconductors with magnetic field have been described by Zawadzki [11] in the frame of Kane's band model The impurity states, attached to a Landau level are classified by quantum numbers for the Landau level n, the angular momentum component M, the bound state β, and spin s. One-photon transitions between impurity states are possible without or with change of the internal quantum number β (see Fig 4) In the former case (Δβ = 0) the transition connects different Landau levels, while in the latter case (Δβ ≠ 0) the transition takes place between different impurity levels attached to the same Landau level. These transitions can be

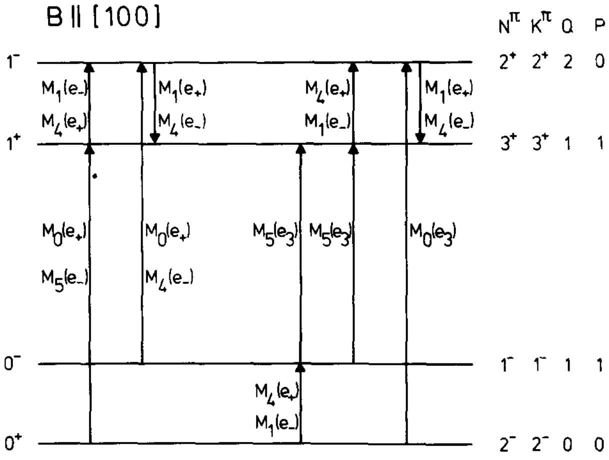


Fig. 3 Two-photon cyclotron and combined resonance. Virtual transitions are characterized by the weights M_i [8].

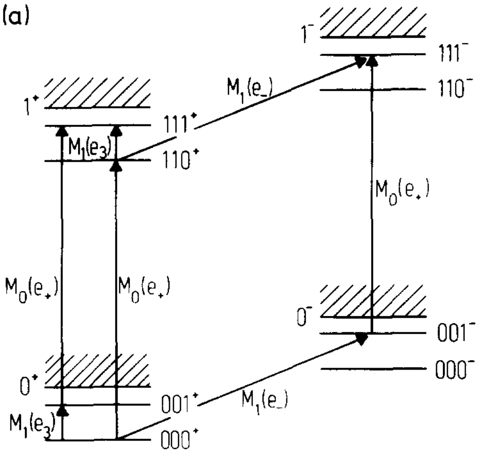
classified as M_0 and M_1, respectively Transitions of types M_2. .M_5 do not occur in this model, which does not consider terms of cubic or tetrahedral symmetry in the Hamiltonian. Possible two-photon transitions between impurity states are shown in Fig 4

V Conclusions

In summary two-photon intraband transitions in the far infrared spectral range have been observed in InSb. Two photon cyclotron resonance and combined resonance could clearly be identified in agreement with two-photon selection rules In an improved experimental arrangement e.g. using thinner samples and applying well defined polarization configurations excited shallow impurity states not accessible to conventional one-photon spectroscopy might be resolved

We thank H R. Trebin for valuable discussions

(a)



(b)

	Δs	ΔM	Δn	Δβ	
e_3	0	0	0	±1	M_1
	-1	+1	+1	0	M_0
	+1	-1	-1	0	M_0
e_+	0	+1	+1	0	M_0
	+1	0	0	±1	M_1
e_-	0	-1	-1	0	M_0
	-1	0	0	±1	M_1

Fig 4 Two-photon shallow donor transitions deduced from electric dipole selection rules after Zawadzki [11]

References

[1] T. Murotani and Y Nisida, J Phys Soc Japan 32, 986 (1972)

[2] E Gornik, T.Y. Chang, T J Bridges, V T Nguyen, I D, McGee, and W Muller, Phys Rev. Letters 40, 1151 (1978)

[3] K Muro, N Yutani, and Sh Narita, J Phys Soc. Japan 49, 593 (1980)

[4] C R Pidgeon, A Vass, G R. Allan, W Prettl, and L Eaves, Phys Rev Letters 50, 1309 (1983)

[5] W Prettl, A. Vass, G R. Allan, and C.R. Pidgeon, Int. J. Infrared and Millimeter Waves 4, 561 (1983).

[6] F Keilmann, 7th Int. Conf. on Infrared and Millimeter Waves, Marseille 1983.

[7] W. Bohm, E. Ettlinger, and W. Prettl, Phys. Rev. Letters 47, 1198 (1981)

[8] H.R. Trebin, U. Rossler, and R. Ranvaud, Phys. Rev. B 20, 686 (1979).

[9] R. Kaplan in Lecture Notes in Physics, vol. 133, p. 138, ed. W. Zawadzki (Springer, Berlin - Heidelberg - New York, 1980).

[10] M. Braun, U. Rossler, J. Phys. C, in print.

[11] W Zawadzki in Theoretical Aspects and New Developments in Magneto-Optics, Nato Advanced Study Institutes Series, Plenum Press 1980, p 347.