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Photoconductivity Studies of Two-Photon Magnetoabsorption in InSb and PbTe

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The photoconductivity of InSb and PbTe subjected to high-intensity CO_2 laser radiation (10.6 and 9.6 μm) has been examined as a function of magnetic field. Sharp maxima have now been observed for laser polarizations both perpendicular and parallel to the magnetic field. These maxima are interpreted as two-photon interband magneto-optical resonances, using a recent extension of the Keldysh ionization theory, rather than the conventional perturbation theory (which predicts maxima only for the perpendicular polarization).

I. INTRODUCTION

WE have completed an experimental and theoretical study of two-photon magnetoabsorption in the small-gap semiconductors InSb and PbTe. The experiments consisted of studying the photoconductivity resonances, at various values of magnetic field, produced by the high-intensity 10.6- and 9.6-μm radiation from a Q-switched CO_2 laser. A preliminary report of these observations was made by Button *et al.*,[1] including a tentative theoretical identification of the PbTe spectrum observed with the laser polarization transverse to the magnetic field. The experiments have now been carried out with both transverse and longitudinal polarizations, at various intensity ratios of the 10.6-μm to the 9.6-μm radiation, and at various values of dc bias. We have been able to make a good correlation of the InSb results with the rather well-known band structure of this material, and have found that these results as well as many aspects of the PbTe results do not agree with the original interpretation. Consequently, a new theoretical approach was developed, which has been reported in detail elsewhere.[2] In Sec. II we give a summary of both theories, including some corrections we have made to the results of the earlier theory. In Sec. III we outline the experimental procedure and in Secs. IV and V give details of the results for InSb and PbTe.

II. THEORY

The preliminary experimental results[1] were interpreted using a theoretical calculation by Zawadzki, Hanamura, and Lax[3] (ZHL), who extended the perturbation theory of multiphoton absorption as worked out, for example, by Braunstein,[4] to include a magnetic field. They found for materials such as InSb and PbTe where one-photon interband transitions are allowed but the two-photon transition is parity-forbidden, that *resonant* two-photon transitions could occur for a *transverse* polarization ($\mathbf{E}\perp\mathbf{H}$) only. These occur by a combination of one interband with one intraband (i.e., cyclotron resonance) virtual transition, with the net selection rule $\Delta n=\pm 1$ for the magnetic or Landau quantum number n. The virtual intermediate state is an adjacent Landau level in either the conduction or valence band. However, the expression given by ZHL takes into account only the valence band as the intermediate state. When we also include the conduction-band state, for simple parabolic bands the term due to this state nearly cancels that due to the valence-band state, so that the transition rate W is smaller than that given in Ref. 3 by a factor $\sim(\omega_c-\omega_v)^2/\omega^2$. The expression we obtain is, for $\mathbf{E}\perp\mathbf{H}$ and $\Delta n=\pm 1$,

$$W_{\perp\pm}=\left(\frac{eE}{2m\omega}\right)^4\frac{(2\mu)^{1/2}}{8\pi}\left(\frac{eH}{\hbar c}\right)^2\left(\frac{|P_{cv}|(\omega_c-\omega_v)}{\hbar(\omega\mp\omega_c)(\omega\mp\omega_v)}\right)^2 \times\sum_n\begin{Bmatrix}n+1\\ n\end{Bmatrix}\left[2\hbar\omega-(\mathcal{E}_{c,n\pm1}-\mathcal{E}_{v,n})\right]^{-1/2},\quad(1)$$

for simple bands such as the two-band model for PbTe (but for $\omega_c\neq\omega_v$). Here m is the free-electron mass, μ is the reduced mass, ω_c and ω_v are the cyclotron frequencies, and ω and E are the laser frequency and electric field. The rate is proportional to the squares of the laser intensity and the magnetic field H, and displays resonant peaks $\sim(2\hbar\omega-\Delta\mathcal{E})^{-1/2}$ whenever the two-photon energy $2\hbar\omega$ equals an interband transition energy $\Delta\mathcal{E}=\mathcal{E}_{c,n\pm1}-\mathcal{E}_{v,n}$.

The situation is different for InSb, where the valence band consists of light- and heavy-hole bands degenerate at $\mathbf{k}=0$. It turns out that the interband matrix element P_{cv} is allowed not only between states with the same n, but also from the heavy-hole state n to the conduction-

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[1] K. J. Button, B. Lax, M. Weiler, and M. Reine, Phys. Rev. Letters **17**, 1005 (1966).

[2] M. Weiler, M. Reine, and B. Lax, Phys. Rev. **171**, 949 (1968).

[3] W. Zawadzki, E. Hanamura, and B. Lax, Bull. Am. Phys. Soc. **12**, 100 (1967); in Proceedings of the Symposium on Nonlinear Optics, Yerevan, USSR, 1967 (to be published).

[4] R. Braunstein, Phys. Rev. **125**, 475 (1962); R. Braunstein and N. Ockman, *ibid.* **134**, A499 (1964).

band state $n-2$. Because of the n dependence of the intraband matrix element, the effects of the intermediate states do not cancel. We find, for ω_c, $\omega_v \ll \omega$,

$$\begin{Bmatrix} W_{\perp,-1} \\ W_{\perp,-3} \end{Bmatrix} = \left(\frac{eE}{2m\omega}\right)^4 \frac{(2\mu)^{1/2}}{8\pi} \left(\frac{eH}{\hbar c}\right)^2 \left(\frac{|P_{cv}{}^{(-2)}|}{\hbar\omega}\right)^2 \times \sum_n \begin{Bmatrix} [(n+1)^{1/2}-(n-1)^{1/2}]^2 \\ [n^{1/2}-(n-2)^{1/2}]^2 \end{Bmatrix} \times \left[2\hbar\omega - \left(\begin{Bmatrix} \mathcal{E}_{c,n-1} \\ \mathcal{E}_{c,n-3} \end{Bmatrix} - \mathcal{E}_{v,n}\right)\right]^{-1/2}. \quad (2)$$

The light-hole transitions $\Delta n = \pm 1$ and the $\Delta n = +1$ transitions for the heavy hole can occur only by the same process as for simple bands with a much smaller rate given by (1). Thus the lowest-lying transition with an appreciable strength is the $\Delta n = -3$ transition $\{\text{heavy hole}, n=3\} \rightarrow \{c, n=0\}$, since the heavy-hole ladders start at $n=2$. This transition may have been observed in our experiments, in addition to the transitions predicted by the other theory which we will now outline.

An alternative approach to multiphoton absorption was developed by Keldysh,[5] who treated it as the high-frequency limit of the time-dependent tunneling induced by the oscillatory electric field of the laser radiation. This theory has now been modified by us (WZL) to include both longitudinal (**E**‖**H**) and transverse (**E**⊥**H**) magnetic fields.[2] The principal feature of this theory is that since the transition is basically a tunneling process, only one transition matrix element is involved, rather than the two for the perturbation theory. The higher-order transitions take place because the energy of the electrons in the separate bands is regarded as oscillatory in time at the laser frequency (and also twice that frequency), so they are able to absorb energy at the laser frequency and its harmonics. Thus the selection rule generally follows the one-photon rule, in this case $\Delta n = 0$ rather than $\Delta n = \pm 1$ for simple bands, with transitions occurring for *both* transverse and longitudinal polarization. The expression obtained in Ref. 2 for **E**‖**H** is

$$W_{\parallel,0} = \sum_n \left(\frac{e^2E^2}{8\mu\omega^2\mathcal{E}_n}\right)^2 \frac{\omega^2(2\mu)^{1/2}}{16\pi^2} \frac{eH}{\hbar c} e^{(4\hbar\omega-\mathcal{E}_n)/\hbar\omega} \times [2\hbar\omega - (\mathcal{E}_{c,n} - \mathcal{E}_{v,n} + e^2E^2/4\mu\omega^2)]^{-1/2}, \quad (3)$$

where $\mathcal{E}_n \equiv \mathcal{E}_{c,n} - \mathcal{E}_{v,n}$. For **E**⊥**H**

$$W_{\perp,\pm m} = \sum_{n,n'} \left(\frac{e^2E^2}{8\mu\omega^2\mathcal{E}_{n'n}}\right)^2 \frac{\omega^2(2\mu)^{1/2}}{8\pi^2} \frac{eH}{\hbar c} e^{\mathcal{E}_{n'n}/\hbar\omega} \alpha_{n'n} \times [2\hbar\omega - (\mathcal{E}_{c,n'} - \mathcal{E}_{v,n} + e^2E^2/4\mu\omega^2)]^{-1/2}, \quad (4a)$$

where $\mathcal{E}_{n'n} = \mathcal{E}_{c,n'} - \mathcal{E}_{v,n}$, $m = |n'-n|$, and

$$\alpha_{n'n} = \left(\frac{1}{m!}\right)^2 \left(\frac{n'!}{n!}\right)^{\pm 1} \left(\frac{\omega_c+\omega_v}{2\omega}\right)^m \times \exp\left(\frac{2(n-n')(\omega_c-\omega_v)}{\omega}\right), \quad (4b)$$

with the upper sign for $n' > n$ and the lower for $n' < n$. Note that the $\Delta n = 0$ selection rule breaks down slightly for **E**⊥**H**. This is because the oscillating electric field couples the neighboring Landau levels so that $\Delta n = \pm m$ transitions are allowed.[6] However, they are weaker than $\Delta n = 0$ by the factor $[(\omega_c+\omega_v)/2\omega]^m \sim H^m$, which is much smaller than 1. The $\Delta n = \pm 1$ transition rate, from Eqs. (4), is then proportional to H^2, as are the transition rates from the perturbation theory [Eqs. (1) and (2)].

We have made an order-of-magnitude calculation from the above expressions and find that for InSb at 100 kOe the $\Delta n = -3$ heavy-hole transitions from Eq. (2) are about an order of magnitude weaker than the $\Delta n = 0$ (and -2) transitions from Eqs. (4) and are comparable in strength to the $\Delta n = \pm 1$ (and -3) transitions from Eqs. (4). For PbTe, the $\Delta n = \pm 1$ transitions from (1) are several orders of magnitude weaker than the $\Delta n = 0$ transitions from (3) and (4). Thus the strong transitions for both materials should be of the tunneling type, following the one-photon selection rules, with the possibility of weak $\Delta n = \pm 1$, -3 transitions in InSb. These predictions have generally been confirmed by the experiments.

III. EXPERIMENTS

The experiments utilized a Q-switched CO_2 laser with a single Brewster window, emitting 1–3-kW peak power, multimode, at both 10.6 and 9.6 μm. The samples were mounted on the cold finger of a liquid-helium Dewar (sample $T \sim 15°$K) so that for both InSb and PbTe, a $\langle 100 \rangle$ axis was parallel to the Dewar axis which was suspended in the bore of a radial-access Bitter solenoid, parallel to the magnetic field. The laser beam was directed along the radial-access bore and focused onto the sample using an aspheric Irtran II lens. The laser polarization was adjusted to be either parallel or perpendicular to the magnetic field by rotating the Brewster window. The polarization in both cases was along $\langle 100 \rangle$ axes so as to eliminate second-harmonic generation in InSb which would have complicated the results. (However, Gibson *et al.*[7] have recently shown that, for zero magnetic field at least, the effects of second-harmonic generation are negligible compared to two-photon absorption for any polarization.)

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[5] L. V. Keldysh, Zh. Eksperim. i Teor. Fiz. **47**, 1945 (1964) [English transl.: Soviet Phys.—JETP **20**, 1307 (1965)].

[6] The coupling is closely related to that obtained by A. G. Aronov, Fiz. Tverd. Tela **5**, 552 (1963) [English transl.: Soviet Phys.—Solid State **5**, 402 (1963)], for a dc electric field.

[7] A. F. Gibson, M. J. Kent, and M. F. Kimmitt, Brit. J. Appl. Phys. **1**, 149 (1968).

The samples were biased with a small electric field, parallel to the magnetic field, and the photoconductivity pulses were detected as changes in the sample voltage using a standard constant-current circuit. The photoconductive signal was filtered, amplified, then processed by a Princeton Applied Research boxcar integrator and displayed on a chart recorder as the magnetic field was swept. Data were taken at various intensity ratios of the 9.6- to the 10.6-μm radiation using filters and also varying the laser pressure and discharge current.

It was found that the magnetic field positions of the photoconductivity maxima varied slightly as a function of the sample bias voltage. The final positions were determined at very slight or zero bias, while the data reported in Ref. 1 were taken at considerably larger bias, so that there is some disagreement between those results and the final results reported here.

IV. InSb RESULTS

In Fig. 1 we present some representative experimental data for InSb, along with the theoretical identification of the experimental peaks. In the lower traces we show the photoconductivity as a function of magnetic field, for both the longitudinal ($\mathbf{E}\|\mathbf{H}$) and transverse ($\mathbf{E}\perp\mathbf{H}$) polarization. The upper curves are the low-lying transition energies for InSb from the one-photon data of Pidgeon and Brown,[8] for $\mathbf{H}\|\langle 100\rangle$. The solid lines (2–4 and 6) occur for $\mathbf{E}\perp\mathbf{H}$, and the dashed lines (1 and 5) for $\mathbf{E}\|\mathbf{H}$. The strong $\mathbf{E}\perp\mathbf{H}$ experimental peaks are repeated in the upper part as error bars at energies corresponding to their identification as absorption of two 9.6-μm photons or of one at 9.6 and one at 10.6 μm. The agreement is excellent and the identifications were roughly confirmed by the relative intensity changes as the intensity at 9.6 μm was varied relative to that at 10.6 μm. The peak intensities also approximately confirm the polarization properties of the theoretical transitions: The peaks at 35 and 73 kOe, which should occur only for $\mathbf{E}\perp\mathbf{H}$, are relatively weak in the $\mathbf{E}\|\mathbf{H}$ trace. We interpret the remaining intensity as being due to inaccuracy in the orientation of the laser polarization.

In addition to these strong transitions there are three weaker peaks (*a*–*c* in the upper trace) which occur at fields corresponding to the two $\Delta n=-3$ heavy-hole transitions (*a* and *c*) and a $\Delta n=-1$ light-hole transition (*b*). The presence of the light-hole transition (stronger than the heavy-hole ones) indicates that these are probably due to the breakdown of the $\Delta n=0, -2$ selection rule in Eqs. (4) rather than to the perturbation mechanism [Eq. (2)]. Thus the InSb results can be explained by the tunneling theory rather than the perturbation theory, which of course gives no explanation of the $\mathbf{E}\|\mathbf{H}$ peaks.

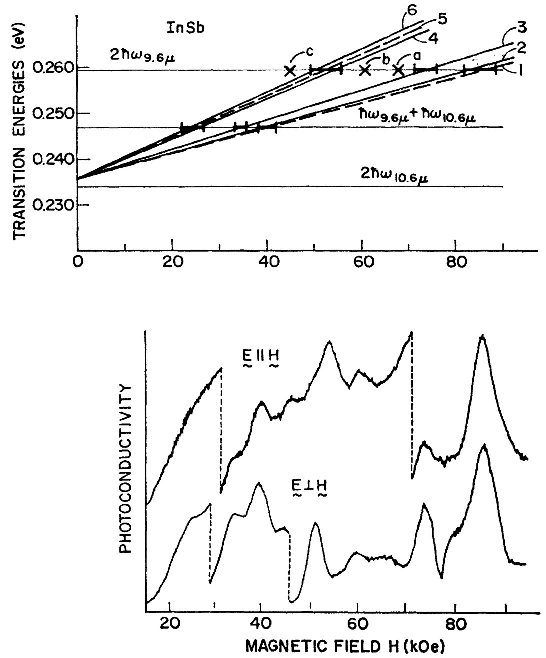

FIG. 1. Multiphoton magnetoabsorption in InSb. Lower curves: experimental traces for two polarizations. Upper curves: theoretical transitions from Ref. 8, with $\mathbf{E}\perp\mathbf{H}$ experimental peaks. Solid lines ($\mathbf{E}\perp\mathbf{H}$, $\Delta n=0, -2$): 2, $a^+(0)a^c(0)$; 3, $a^-(2)a^c(0)$; 4, $b^-(2)b^c(0)$; 6, $b^+(0)b^c(0)$. Dashed lines ($\mathbf{E}\|\mathbf{H}$, $\Delta n=0, -2$): 1, $b^-(2)a^c(0)$; 5, $a^+(0)b^c(0)$. Crosses ($\mathbf{E}\perp\mathbf{H}$, $\Delta n=\pm 1, -3$): *a*, $a^-(3)a^c(0)$; *b*, $a^+(1)a^c(0)$; *c*, $b^-(3)b^c(0)$. See Ref. 8 for notation.

V. PbTe RESULTS

An interpretation of the PbTe results has been made along the lines discussed for InSb. However, for PbTe an accurate and complete model of the band-edge structure is not yet available. The most recent experimental work is that of Aggarwal, Smith, and Lax,[9] who have measured the one-photon transition energies for $\mathbf{E}\perp\mathbf{H}$ using the magnetopiezoreflection technique. They have determined the reduced-effective-mass parameters and shown that the light-mass transitions can be described reasonably well by a two-band nonparabolic model.[10] We have calculated both $\Delta n=0$ and $\Delta n=\pm 1$ transition energies using this model, using the mass values of Ref. 9 for the [100] orientation ($\mu=0.018m_0$), but varying the energy gap to obtain a best fit to our experiments. (The energy gap is known to vary with temperature and also with stress, which was present in the experiments of Ref. 9, where the gap was $\mathcal{E}_g=0.019$ eV.)

In Fig. 2 we show the $\Delta n=0$ fit along with a representative experimental trace for $\mathbf{E}\perp\mathbf{H}$. (A similar spectrum was observed for $\mathbf{E}\|\mathbf{H}$, i.e., no spin splitting was

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[8] C. R. Pidgeon and R. N. Brown, Phys. Rev. **146**, 575 (1966).

[9] R. L. Aggarwal, U. Smith, and B. Lax, in Proceedings of the International Conference on the Physics of Semiconductors, Moscow, 1968 (to be published); U. Smith, R. Aggarwal, and B. Lax, Bull. Am. Phys. Soc. **13**, 429 (1968).

[10] B. Lax, J. G. Mavroides, H. J. Zeiger, and R. J. Keyes, Phys. Rev. Letters **5**, 241 (1960).

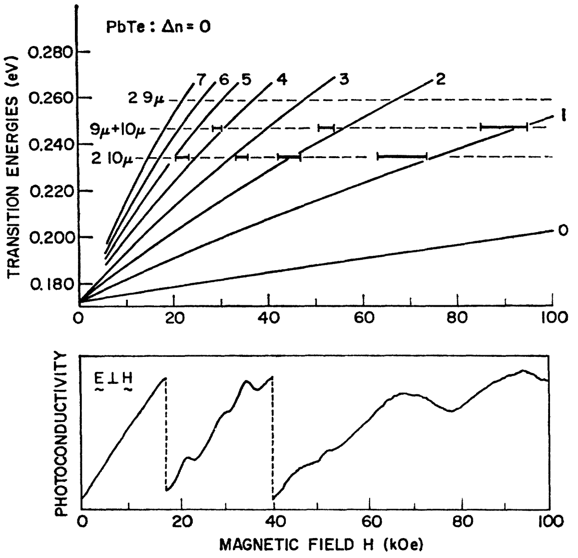

FIG. 2. Multiphoton magnetoabsorption in PbTe. Lower curve: experimental trace for E⊥H. Upper curves: theoretical transitions calculated from the two-band nonparabolic model, with the one-photon selection rule $\Delta n=0$, labeled by the n of the initial and final states.

observed. A similar result was reported in one-photon magnetoabsorption experiments by Mitchell *et al.*,[11] although Butler and Calawa[12] did report the observation of a spin splitting in recombination radiation.) Notice that the peaks are much broader than for InSb. This is because PbTe is not available in nearly as pure form as InSb. (Our samples of PbTe had $n\approx10^{16}/\text{cm}^3$ and $10^{17}/\text{cm}^3$, while the InSb had $n\approx10^{14}/\text{cm}^3$.) Consequently, a large range of band parameters could be used to fit the data, so that we do not regard these experiments as providing any measure of these parameters.

The experimental peaks are recorded as error bars in the upper half of Fig. 2 according to their identification as two 10.6-, two 9.6-, or 9.6- plus 10.6-μm absorptions. The agreement is reasonable, although not as good as the InSb identification. In fact, the data can also be fitted by a $\Delta n=\pm1$ identification as shown in Fig. 3, where the 40-kOe peak has now been identified as a 9.6- plus a 10.6-μm peak instead of two 10.6-μm ones. Varying the relative 9.6-μm intensity did not rule out either interpretation, since it was difficult to get an accurate measure of the intensities of such broad peaks relative to the large field-dependent background.

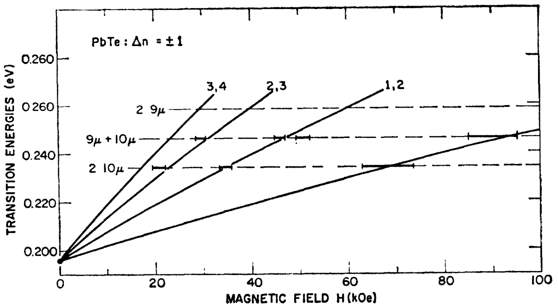

FIG. 3. Upper part of Fig. 2 recalculated for $\Delta n=\pm1$ with a different energy gap and assuming $\omega_c=\omega_v$, labeled by the n of the initial and final states.

It is evident from a comparison of Figs. 2 and 3 that in fact the $\Delta n=\pm1$ identification appears slightly better than $\Delta n=0$. However, the $\Delta n=\pm1$ theory provides no explanation for the E||H results, and furthermore is not consistent with the InSb identification and with the order-of-magnitude calculations in Sec. II. We were able to make a clear $\Delta n=0$ interpretation of one PbTe spectrum taken with a warm sample (~50°K) where the gap was somewhat larger so that fewer lines were observed. Thus from an over-all point of view we reject the $\Delta n=\pm1$ perturbation theory in favor of the $\Delta n=0$ tunneling theory.

VI. DISCUSSION

The photoconductivity spectra of InSb and PbTe at 10.6 and 9.6 μm can be interpreted in terms of the two-photon absorption mechanism originally described by Keldysh[5] and recently extended by WZL to describe resonant magnetoabsorption in a magnetic field. This method of detection, in which only the magnetic field can be varied and not one of the photon frequencies, has inherent limitations, especially since in photoconductivity measurements the intensity and line-shape information is only approximate. It is hoped that eventually the two-photon technique,[13] in which a variable frequency is employed, can be applied to InSb and PbTe in a magnetic field so that the predictions of the tunneling theory can be treated more quantitatively.

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[11] D. L. Mitchell, E. D. Palik, and J. N. Zemel, in *Proceedings of the Seventh International Conference on the Physics of Semiconductors, Paris, 1964* (Academic Press Inc., New York, 1965), p. 325.

[12] J. F. Butler and A. R. Calawa, in *Physics of Quantum Electronics*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Co., New York, 1966), p. 458.

[13] J. J. Hopfield and J. M. Worlock, Phys. Rev. **137**, A1455 (1965).