

## MULTIPHOTON MAGNETO-OPTICAL RESONANCE IN PbTe AND InSb

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High intensity magnetic fields are used to observe multiphoton direct transitions in PbTe and InSb. These are resonant absorptions of high-intensity CO<sub>2</sub> laser radiation by means of higher order transitions of the type proposed by Braunstein.

We have observed photoconductivity resonances and recombination radiation in PbTe and InSb due to multiple-photon interband transitions between Landau levels in magnetic fields up to 100 kG. The magnetic field dependence provides the first direct evidence of resonant multiphoton absorption, and, in principle allows one to distinguish among two-, three-, and four-photon processes and to study the relative intensities of the different processes in different crystal symmetries. The incident radiation of 10.6- and 9.6- $\mu$  wavelengths from a Q-switched CO<sub>2</sub> laser was focused on the samples, using an apparatus similar to that described by Patel *et al.*<sup>1</sup> in their recent observations of multiphoton plasma production. The wavelengths of 10.6 and 9.6  $\mu$  correspond to photon energies of 0.117 and 0.124 eV, respectively, which are too small to cause one-photon interband transitions in these materials.

Figures 1 and 2 show the photoconductivity as a function of applied magnetic field intensity in the two materials. The sharp peaks correspond to multiphoton absorptions due to direct interband transitions between Landau sub-bands at the peak of the density of states. The problem of multiphoton absorption with no magnetic field has been treated by Braunstein<sup>2</sup> using higher order time-dependent perturbation theory. Zawadzki, Hanamura, and Lax<sup>3</sup> have extended this theory to the magnetic field case. If the absorptions were due to allowed transitions between two simple parabolic bands, neglecting spin, we would expect to see resonances satisfying the following relation:

$$\hbar\omega = \mathcal{E}_g + (n_1 + \frac{1}{2})\hbar\omega_{c1} + (n_2 + \frac{1}{2})\hbar\omega_{c2}, \quad (1)$$

where  $\hbar\omega$  is the multiphoton energy,  $\mathcal{E}_g$  is the energy gap, the  $n_i$  are the magnetic quantum numbers,  $\omega_{ci} = eH/m_i$  the cyclotron frequencies,  $m_i$  are the effective masses of hole and electron, and  $H$  is the magnetic field intensity. A theoretical treatment for PbTe and InSb, however, requires Eq. (1) to be modified to include nonparabolicity of bands, the influ-

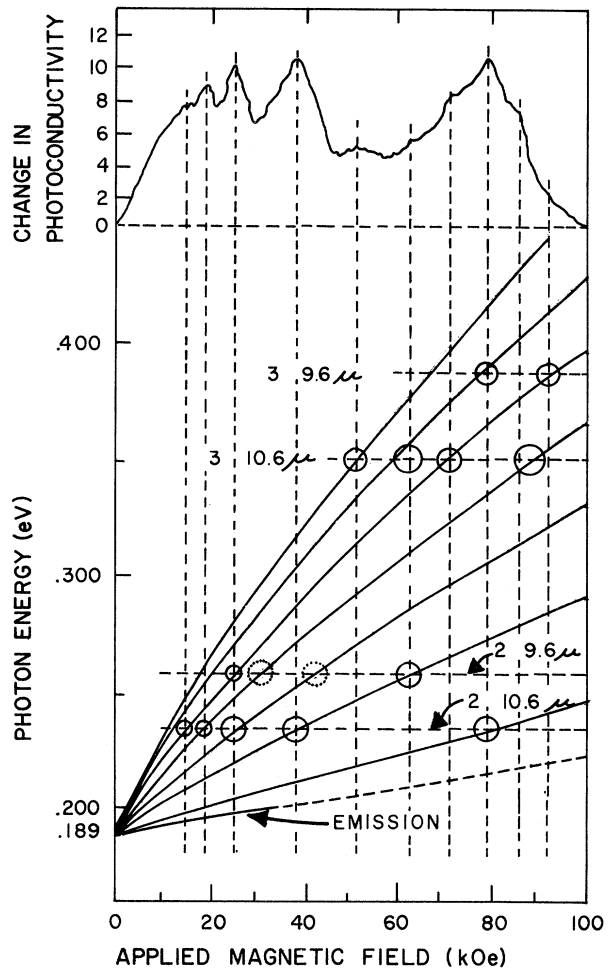


FIG. 1. The photoconductivity peaks of PbTe (upper trace) due to multiphoton absorption are compared with transitions calculated from a simple two-band non-parabolic model for selection rules  $\Delta n = \pm 1$ . The best fit was obtained by using  $\mathcal{E}_g = 0.189$  eV and reduced effective mass  $m^* = 0.018$ , which were obtained from the magnetic field dependence of the observed spontaneous emission. The extrapolation of the field dependence of the emission is shown as a dashed line. The horizontal dashed lines indicate integral multiples of the energies of the incident photons from a 1-kW Q-switched CO<sub>2</sub> laser emitting at wavelengths of 10.6 and 9.4  $\mu$ , polarized with  $E \perp H \parallel \langle 100 \rangle$ . The dotted circles were observed by filtering out the 10.6- $\mu$  radiation. The sample temperature was  $\approx 20^\circ\text{K}$ .

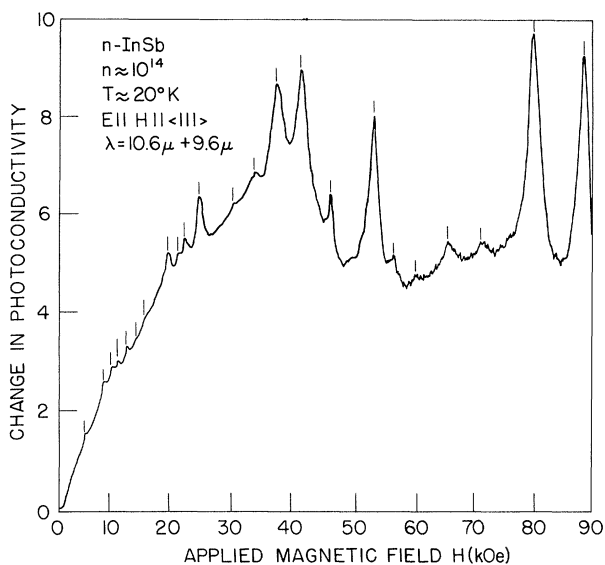


FIG. 2. The photoconductivity peaks of InSb attributed to multiphoton interband absorption.

ence of other bands, and spin splitting.

For the case of PbTe,<sup>4</sup> the strong peaks in the photoconductivity shown in Fig. 1 fall on a straight line when plotted against  $1/H$ , the line extrapolates to  $n=0$ , but the points correspond to integral steps which indicates that the selection rule for the transition is  $\Delta n = \pm 1$ . This has been predicted for two-photon excitations between bands of opposite parity, such as in PbTe, by Zawadzki, Hanamura, and Lax,<sup>3</sup> by considering the transition as a combination of interband-intraband virtual transitions. Using this selection rule, we calculated the curves in the lower part of Fig. 1 using the two-band nonparabolic model<sup>5</sup> and values of the energy gap and effective mass of  $\mathcal{E}_g = 0.189$  eV and  $m^* = 0.018$ . Wherever one of these curves crosses any of the horizontal dashed lines corresponding to the energies of multiple incident photons, we see a peak in the photoconductivity trace. The two dotted circles indicate peaks which have been observed by filtering out the  $10.6\text{-}\mu$  component of the laser beam using a  $\text{CaF}_2$  window. The photoconductivity trace for  $E \parallel H$  was essentially the same as for  $E \perp H$ . This was also observed by Mitchell, Palik, and Zemel<sup>6</sup> for one-photon magnetoabsorption.

Although Mitchell, Palik, and Zemel<sup>6</sup> reported  $\mathcal{E}_g = 0.19$  eV and  $m^* = 0.021$ , our values were obtained from the magnetic field dependence of the emission of recombination radiation,

which has been plotted as a dashed line on the lower part of Fig. 1. The intensity of this emission decreased rapidly with increasing magnetic field. The intercept and the slope of this line give our values for gap and mass using the selection rule  $\Delta n = 0$  for emission. Butler and Calawa<sup>7</sup> have also reported the magnetic field dependence of emission from PbTe, which is essentially the same. The slope of the line corresponding to their unpolarized emission yielded a mass of 0.022, and they reported an energy gap at  $10^\circ\text{K}$  of 0.188 eV. They also observed emission polarized parallel to the magnetic field from which they were able to determine a  $g$  factor of 29. We have not yet attempted to confirm the observation of emission in the  $\pi$  configuration.

For the case of InSb, our preliminary data at low laser power correlated approximately with the one-photon,  $\Delta n = 0, -2$  transitions observed by Pidgeon and Brown.<sup>8</sup> However, the more distinct peaks observed with higher laser power, as shown in Fig. 2, do not correlate exactly with the  $\Delta n = 0, -2$  transitions. The prediction<sup>3</sup> for InSb is similar to that for PbTe in that the two-photon transitions should obey " $\Delta n = \pm 1$ ", i.e.,  $\Delta n = \pm 1, -3$ , and the three-photon transitions should be  $\Delta n = 0, -2$ . However, there is the possibility that the observed peaks could be due to carriers produced by the resonant absorption of single photons from the second harmonic generation process, which can be occurring simultaneously with multiphoton absorption in InSb, but not in PbTe. Thus a great number of transitions are possible, and a program is under way to attempt to identify in detail the observed peaks.

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<sup>3</sup>W. Zawadzki, E. Hanamura, and B. Lax, to be published.

<sup>4</sup>Sample kindly supplied by A. C. Beer, Battelle Memorial Institute, sample No. 19733-21D.

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<sup>7</sup>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 Company, Inc., New York, 1966), p. 458.

### TEMPERATURE AND STRESS DEPENDENCE OF ELECTRON LIFETIME IN *p*-TYPE SEMICONDUCTORS BETWEEN 1.5 AND 4.2°K

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A low-temperature electron-lifetime study has been carried out by means of linewidth and intensity measurements in a cyclotron-resonance experiment for two kinds of uncompensated *p*-type materials, namely Si/B and Ge/Zn. It is found that in these materials, the carrier recombination time is much shorter than the thermalization time near 1.5°K—an important prerequisite for achieving such electron population inversion within the conduction band as suggested by Wolff for the case of InSb.<sup>1</sup> Another quite unexpected finding is that application of uniaxial stress drastically increases the electron lifetime. Analysis of the electron-resonance signals with and without stress shows that the main acceptor itself is responsible for such a rapid recombination time. Since the cyclotron-resonance technique in itself provides a powerful tool for studying carrier lifetime because of its ability of clearly distinguishing electron and hole signals, the present method certainly furnishes a most direct determination of the electron-capture rate by a neutral acceptor as a function of temperature and stress.

In an earlier paper<sup>2</sup> we presented a cyclotron-resonance study of electron scattering by neutral group-III acceptors in germanium, and saw that the results may, at least qualitatively, be interpreted in terms of the positron-hydrogen scattering model. In the course of extended study using the materials mentioned in the beginning, the authors have met with singular behavior of the electron resonance: (1) The signal intensity rapidly goes down as the temperature is lowered, and (2) the line-

width becomes unexpectedly broad and decidedly deviates from what is predictable by the positron-hydrogen scattering picture.<sup>2</sup> These facts no doubt manifest a contribution of lifetime broadening to the observed cyclotron-resonance linewidth.

The materials used are (1) boron-doped silicon crystals with  $N_B = 4.3 \times 10^{14}$ ,  $9.5 \times 10^{14}$ , and  $2.8 \times 10^{15} \text{ cm}^{-3}$ , and (2) zinc-doped germanium crystals with  $N_{Zn} = 1.2 \times 10^{14}$  and  $2.1 \times 10^{15} \text{ cm}^{-3}$ . The linewidths measured at 35 Gc/sec with a superheterodyne detection system are given against temperature in Fig. 1 for representative samples. Any spurious broadening arising from carrier heating<sup>3</sup> or carrier-carrier interaction effect<sup>4</sup> is carefully avoided by minimizing both microwave power and light

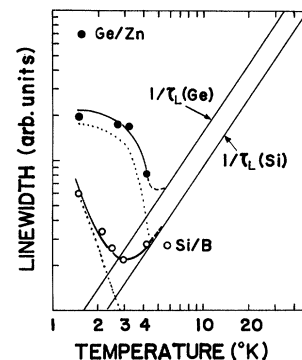


FIG. 1. Cyclotron-resonance linewidths of electron against temperature for representative crystals of Si/B and Ge/Zn. The straight lines denoted by  $1/\tau_L$  correspond to the lattice scatterings for Ge and Si. The dotted lines indicate the contributions of lifetime broadening.