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Phonon Cyclotron Resonance in the Infrared Absorption in n-Type InSb†

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A series of peaks has been observed in the infrared absorption by free carriers in n-type InSb under applied magnetic field. With decreasing field, the peaks converge to an energy equal to that of a LO phonon. The energy separations between the peaks are related to the separations of Landau levels. These peaks are called phonon cyclotron resonances, and they are associated with indirect transitions involving LO phonons. A theory is presented which accounts for various aspects of the observation including the positions and the strengths of the resonances as well as the effect of polarization on the resonances. Some peaks were observed in addition to the phonon cyclotron resonances, which appear to represent the third harmonic of cyclotron resonance and the second harmonic with spin flip.

INTRODUCTION

PHOTON absorption in a crystal can be produced by electron transitions between two levels within one energy band. Normally, such an absorption process is indirect in that electron-lattice interaction must be involved in order to conserve both energy and momentum. In a magnetic field, direct transitions can occur between Landau sub-bands giving cyclotron-resonance absorption. Indirect transitions with the help of lattice interaction are also present. Since the energy spread of optical phonons is small, indirect transitions involving optical phonons might be expected to produce also resonances in the absorption. Unlike the direct cyclotron transition for a simple parabolic energy band, the indirect transitions might occur between any two Landau sub-bands, i.e., the difference Δn of the quantum numbers is not limited to $\Delta n=\pm 1$. Thus the electrons in a Landau sub-band could give rise to a series of resonances in absorption corresponding to transitions to successively higher sub-bands. Such peaks in absorption will be referred to as phonon cyclotron resonances. Evidence for the existence of such absorption peaks in n-type InSb was found by Y. Marfaing[1] in our laboratory. At the same time a peak of this nature was observed by McCombe *et al.*,[2] and recently two observed peaks were reported by Dickey and Johnson[3] in the same material. The observed peaks were attributed to phonon cyclotron resonances only by energy consideration.

We have made measurements over a wide frequency range.[4] A series of seven peaks was observed and studied with varying magnetic field. Polarized radiations have been used with $\mathbf{E}\perp\mathbf{B}$ or $\mathbf{E}\parallel\mathbf{B}$, showing that the peaks were not detectable with $\mathbf{E}\parallel\mathbf{B}$. Hall measurements made on the samples showed that there was no appreciable freeze-out of carriers with the magnetic field used in the optical measurements, thus making sure that the observed resonances were indeed produced by free carriers. The results are examined in the light of the theory presented. A theoretical treatment similar to ours has been reported earlier by Bass and Levinson,[5] predicting the phonon cyclotron-resonance absorption. Our studies were started independently. Those theoretical results which overlap are in agreement. We give explicit expressions for carriers with degenerate distribution, giving peaks for $E\perp B$ which are resaonable compared with the observed resonances. We consider also the case of $E\parallel B$, showing that absence of peaks is expected. The nature of these peaks is thus established. The measurements showed some extra peaks which evidently do not involve phonon participation. These peaks are tentatively attributed to a harmonic cyclotron resonance and a cyclotron resonance with spin flip.

THEORY

The transition is a second-order process involving electron radiation H_R and electron lattice H_L interactions. The transition rate is given by

$$P=\frac{2\pi}{\hbar}\left|\sum_{I}\frac{\langle F|H_R|I\rangle\langle I|H_L|0\rangle}{E_0-E_I}+\sum_{I'}\frac{\langle F|H_L|I'\rangle\langle I'|H_R|0\rangle}{E_0-E_{I'}}\right|^2\rho_E=\frac{2\pi}{\hbar}|M|^2\rho_E, \quad (1)$$

where 0, I, and F refer to the initial, intermediate, and final states, respectively. Each state is specified by a quantum number N_ν of the radiation field, a set of quantum numbers, n_q's, for the phonons, and the quantum numbers n, k_z, k_x, and s for the electron. The magnetic field is applied in the z direction and will be represented by a vector potential $\mathbf{A}=(-By,0,0)$. ρ_E is

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* Now at Xerox Corporation, Xerox Square, Rochester, N. Y.
[1] See Ref. 2.
[2] B. D. McCombe, S. G. Bishop, and R. Kaplan, Phys. Rev. Letters **18**, 748 (1967).
[3] D. H. Dickey and E. J. Johnson, Bull. Am. Phys. Soc. **13**, 430 (1968).
[4] A brief report of some results obtained with unpolarized radiation is contained in a paper by H. Y. Fan, in *Proceedings of the Ninth International Conference on the Physics of Semiconductors, Moscow, 1968* (Publishing House "Nauka," Leningrad, 1968), p. 135.
[5] F. G. Bass and I. B. Levinson, Zh. Eksperim. i Teor. Fiz. **49**, 914 (1965) [English transl.: Soviet Phys.—JETP **22**, 635 (1966)].

the density of final states. The interactions are given by

$$H_R=-\frac{e}{m}\left(\frac{2\pi\hbar^2N_\nu}{\epsilon_1\hbar\omega}\right)^{1/2}\left(\mathbf{p}-\frac{e}{c}\mathbf{A}\right)\cdot\boldsymbol{\eta}, \tag{2a}$$

$$H_L=\sum_{\mathbf{q}}\left(C_{\mathbf{q}}a_{\mathbf{q}}e^{i\mathbf{q}\cdot\mathbf{r}}+C_{\mathbf{q}}^*a_{\mathbf{q}}^\dagger e^{-i\mathbf{q}\cdot\mathbf{r}}\right), \tag{2b}$$

where ϵ_1 is the real dielectric constant, N_ν is the photon density, $\boldsymbol{\eta}$ is the direction of polarization of radiation, $\mathbf{A}$ is the vector potential of the applied magnetic field, and $a_{\mathbf{q}}^\dagger$ and $a_{\mathbf{q}}$ are the phonon creation and destruction operators, respectively.

$$C_q=-i\frac{(2\pi e^2\hbar\omega_0)^{1/2}}{q}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right)^{1/2}$$
$$=-i\frac{(4\pi\alpha)^{1/2}}{q}\left(\frac{\hbar}{2m\omega_0}\right)^{1/4}\hbar\omega_0, \tag{3}$$

where ω_0 and $\mathbf{q}$ are, respectively, the frequency and the wave vector of the phonon.

Figure 1 shows schematically absorption transitions in the electron-energy diagram. In the absence of magnetic field, the states I' and 0 have nearly the same electronic wave vectors, which differ only by $\Delta\mathbf{k}=\mathbf{K}$, and the wave vector $\mathbf{K}$ of the radiation field may be considered negligible. The same applies to the states I and F. In a magnetic field B, radiation polarized with $E\perp B$ gives matrix elements of H_R between two neighboring Landau levels with $\Delta n=\pm1$. The intermediate states of the transitions are indicated by I_1, I_2, and I_1'. Radiation polarized with $E\parallel B$ has matrix elements of H_R for $\Delta n=0$, and the intermediate states of the transitions are indicated by I_3 and I_3'. The diagrams show only the Landau sub-bands of one spin. Radiative transitions with spin inversion and transitions between nonadjacent Landau sub-bands can occur because of spin-orbit coupling together with a nonparabolicity of the energy band or with a lack of inversion symmetry.[6] Absorptions given by these transitions, known, respectively, as combined resonance[2] and harmonic cyclotron resonance,[7] have been observed experimentally in InSb. These transitions are comparatively very weak and need not be considered as giving intermediate states in our problem.

The matrix element of H_L for the interaction with LO phonons has the form

$$\langle n'k_z'k_x',n_{\mathbf{q}}\pm1|H_L|nk_zk_x,n_{\mathbf{q}}\rangle$$
$$=C_q(n_q+\tfrac{1}{2}\pm\tfrac{1}{2})\delta(k_z'\pm q_z,k_z)\delta(k_x'\pm q_x,k_x)$$
$$\times\int_{-\infty}^{+\infty}dy\,e^{iq_yy}\phi_{n'}\left(y+\frac{k_x'}{\sqrt{S}}\right)\phi_n\left(y+\frac{k_x}{\sqrt{S}}\right). \tag{4}$$

[6] E. I. Rashba and V. I. Sheka, Fiz. Tverd. Tela 3, 1735 (1961) [English transl.: Soviet Phys.—Solid State 3, 1527 (1961)]; V. I. Sheka, Fiz. Tverd. Tela 6, 3099 (1964) [English transl.: Soviet Phys.—Solid State 6, 2470 (1965)]; R. L. Bell and K. T. Rogers, Phys. Rev. 152, 746 (1966).

[7] E. D. Palik and R. F. Wallis, Phys. Rev. 130, 41 (1963).

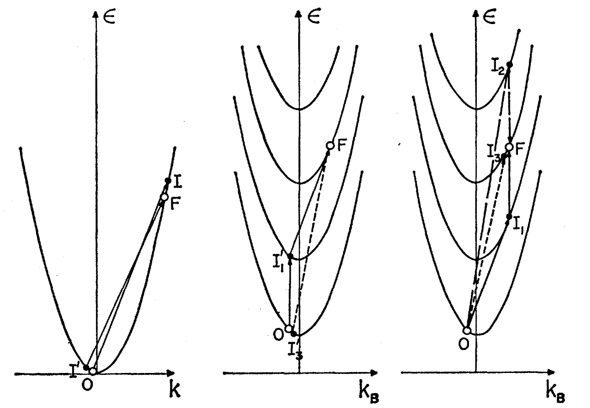

FIG. 1. Electron-energy diagram showing phonon-assisted optical transitions. The left-hand diagram is for $B=0$. The other two diagrams are for the case with an applied magnetic field.

The upper sign is for phonon creation and the lower sign for phonon annihilation. In the free-particle model, ϕ is a harmonic-oscillator wave function and the integral has the expression

$$(-1)^{n-n'}e^{\pm iq_xq_y/2S}\left(\frac{n'!}{n!}\right)^{1/2}$$
$$\times e^{-q_\perp^2/4S}\left(\frac{q_\perp}{\sqrt{2S}}\right)^{n-n'}L_{n'}^{n-n'}\left(\frac{q_\perp^2}{2S}\right) \tag{5}$$

for $n\geq n'$, where $q_\perp^2=q_x^2+q_y^2$, $S=eB/c\hbar$, and $L_{n'}^{n-n'}$ is the associated Laguerre polynomial. In contrast to the case of H_R, the matrix element exists between states of any n and n', and is not restricted to adjacent Landau sub-bands. There are also nonvanishing matrix elements between states of opposite spins[8] because of interactions involving spin which are not included in (2b). The effect of these should be small in our problem. Consider the excitation of an electron from the state (n,k) to the state $(n',k-q)$ with phonon creation. The condition of energy conservation is

$$\hbar\omega-\hbar\omega_0-(\epsilon_{n'}-\epsilon_n)-(\hbar^2/2m)(q_z^2-2k_zq_z)$$
$$=\hbar\Omega_{n'n}-(\hbar^2/2m)(q_z^2-2k_zq_z)=0, \tag{6}$$

where ω is the frequency of radiation and $\epsilon_{n'}$ and ϵ_n are the energies of the Landau levels. With a fixed $q_\perp$ of the emitted phonon, (6) is satisfied at two values of q_z for a given $\hbar\Omega_{n'n}$:

$$q_{1,2}=k_z\pm[(2m/\hbar^2)\hbar\Omega_{n'n}+k_z^2]^{1/2}. \tag{7}$$

At each value of q_z, the density of states for the transitions is given by

$$\rho=(1/2\pi)(m/2\hbar^2)^{1/2}(\hbar\Omega_{n'n}+\hbar^2k_z^2/2m)^{-1/2}. \tag{8}$$

Consider first radiation polarized with $\mathbf{E}\perp\mathbf{B}$. Nonvanishing matrix elements of H_R exist between adjacent

[8] I. M. Tsidilkovskii, M. M. Akselrod, and S. I. Uritsky, Phys. Status Solidi 12, 667 (1965).

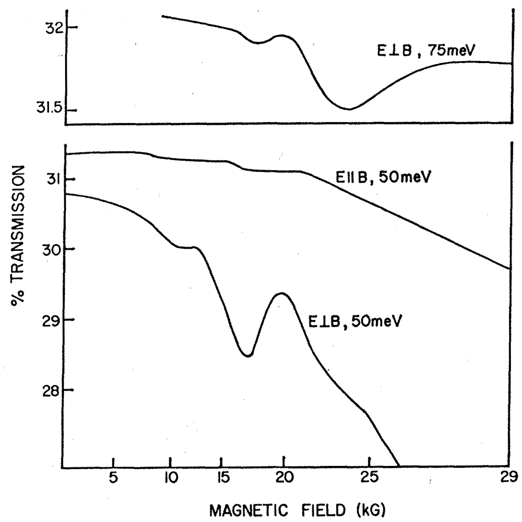

FIG. 2. Transmission as a function of applied magnetic field for n-type InSb with a carrier concentration of $n=1.04\times10^{16}$ cm^{-3}. The photon energy and the polarization used are given for each curve. The sample thickness is 1.5 mm.

Landau sub-bands $\Delta n=\pm1$ which are given by

$$|\langle n+1|H_R|n\rangle| = (e\hbar/m)(2\pi N_\nu/\epsilon_1\hbar\omega)^{1/2}(\tfrac{1}{2}m\hbar\omega_c)^{1/2}(n+1)^{1/2}, \quad (9)$$

where N_ν is the photon density of radiation, ω_c is the cyclotron frequency, and ϵ_1 is the refractive index squared. Integrating (1) over the $\mathbf{q}_\perp$ space, we get the probability of the excitation of the $(n\mathbf{k})$ electron to the n' sub-band with the creation of any phonon:

$$P_{n',n\mathbf{k}}=\rho\frac{2\pi}{\hbar}\frac{1}{4\pi^2}\left(\int q_\perp dq_\perp|M|^2{}_{q_z=q_1} + \int q_\perp dq_\perp|M|^2{}_{q_z=-q_2}\right). \quad (10)$$

The integrand contains a factor

$$1/q^2=1/(q_\perp^2+q_z^2), \quad (11)$$

which is brought in by the coefficient C_q given by (4). As we shall see, resonance absorption occurs at $\hbar\Omega=0$. Near the resonance, the value of q_z is of the order of k_z. Most of the electrons have a small value of k_z owing to the large density of states at the Landau levels. Furthermore, in a degenerate distribution, k_z is limited by the Fermi energy ζ; $k_z\lesssim k_\zeta=(2m\zeta/\hbar^2)^{1/2}$. This is to be compared with the range of $q_\perp$ giving important contribution to the integral (10), which is of the order of $(2S)^{1/2}=(2m\hbar\omega_c/\hbar^2)^{1/2}$, as can be seen from (4) and (5). Under conditions suitable for the study of resonance, $k_\zeta^2\ll2S$. By neglecting q_z^2 in the factor (11), the integration in (10) can be carried out, and we get the following expression for the absorption coefficient contributed by the electron:

$$K_{n',n\mathbf{k}}=[(\sqrt{\epsilon_1})/cN_\nu]P_{n',n\mathbf{k}} =A(\omega,\omega_c)(\hbar\Omega_{n'n}+\hbar^2k_z^2/2m)^{-1/2}, \quad (12)$$

$$A(\omega,\omega_c)=\alpha\frac{\pi}{c\sqrt{\epsilon_1}}\frac{\hbar e^2}{m}\frac{(\hbar\omega_0)^{1/2}}{\hbar\omega}\times\left(\frac{\omega_0\omega_c}{(\omega-\omega_c)^2}+\frac{\omega_0\omega_c}{(\omega+\omega_c)^2}\right). \quad (13)$$

This result agrees with that obtained by Bass and Levinson.[5]

We now sum up the contribution of all electrons in a sub-band n. For a degenerate distribution valid for low temperatures, we get

$$K_{n'n}=N_nA(\omega,\omega_c)(2\zeta^{1/2})^{-1}\times\{\ln[2(\zeta^2+\zeta\hbar\Omega_{n'n})^{1/2}+2\zeta+\hbar\Omega_{n'n}]-\ln(\hbar\Omega_{n'n})\}, \quad (14)$$

where ζ is the Fermi energy measured from the edge of nth Landau band and N_n is the concentration of electrons in the sub-band n. At

$$\hbar\Omega_{nn'}=\hbar\omega-\hbar\omega_0-(\epsilon_{n'}-\epsilon_n)=0, \quad (15)$$

the absorption has a singularity which is the phonon cyclotron resonance. With increasing ω, a resonance occurs every time (15) is satisfied with a larger n'. The radiation used in measurements has a finite frequency spread, and the quantity of interest is the average absorption over a frequency range. Expression (14) averaged over a range $\hbar\Delta\omega$ has a maximum given by

$$(\bar{K}_{n'n})_m=N_nA(\omega,\omega_c)(1/2\zeta^{1/2})[\ln(4\zeta/\hbar\Delta\omega)+1], \quad (16a)$$

provided $\hbar\Delta\omega\ll\zeta$. In case ζ is very small, we get, by neglecting k_z in (12),

$$(\bar{K}_{n'n})_m=2N_nA(\omega,\omega_c)/(\hbar\Delta\omega)^{1/2}. \quad (16b)$$

Consider now radiation polarized with $\mathbf{E}\parallel\mathbf{B}$. We mentioned previously that nonvanishing matrix elements of H_R exist only for $\Delta n=0$. The transition via intermediate I_3' (see Fig. 1) involves

$$|\langle n|H_R|n\rangle|=(e\hbar/m)(2\pi N_\nu/\epsilon_1\hbar\omega)^{1/2}\hbar k_z, \quad (17)$$

and the transition via I_3 involves

$$|\langle n'|H_R|n'\rangle|=(e\hbar/m)(2\pi N_\nu/\epsilon_1\hbar\omega)^{1/2}\hbar(k_z-q_z). \quad (18)$$

The combined contribution of these two processes gives to the factor $|M|^2$ of (1) an additional q_z^2 dependence. Referring to (10), (12), and (7), we see that the dependence of $K_{n',n\mathbf{k}}$ on k_z is now given by

$$K_{n',n\mathbf{k}}\propto(\hbar^2/2m)(q_1^2+q_2^2)(\hbar\Omega_{n'n}+\hbar^2k_z^2/2m)^{-1/2} =(\hbar\Omega_{n'n}+\hbar^2k_z^2/m)(\hbar\Omega_{n'n}+\hbar^2k_z^2/2m)^{-1/2}. \quad (19)$$

Summation over all electrons in the sub-band n gives

$$K_{n'n}=B_{n'n}(\omega,\omega_c)N_n(\hbar\Omega_{n'n}+\zeta)^{1/2}. \quad (20)$$

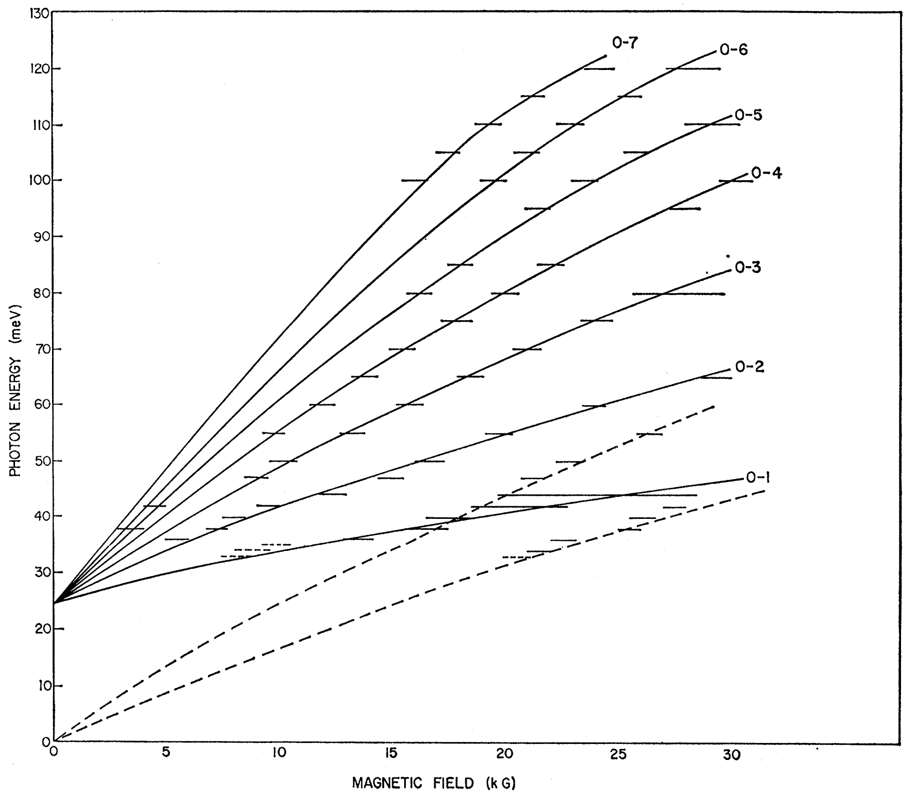

FIG. 3. Photon energy of minima in transmission is plotted against magnetic field. Data were taken with $E\perp B$. Only the three dotted points were taken with unpolarized radiation. The solid curves are drawn through the experimental data, converging to $\hbar\omega_0$ at $B=0$. The dashed curves are drawn parallel to the 0-3 and 0-2 curves, respectively.

The absorption begins at a frequency corresponding to

$$\hbar\Omega_{n'n}=-\zeta, \tag{21}$$

and there is no singularity which would give a resonance peak. For electrons in the lowest Landau sub-band, $n=0$, we get

$$B_{n'0}(\omega,\omega_c)=\alpha\frac{\pi}{c\sqrt{\epsilon_1}}\frac{\hbar e^2}{m}\frac{(\hbar\omega_0)^{1/2}}{(\hbar\omega)^2}\frac{\omega_c\omega_0}{\omega^2}\frac{1}{n'+1}. \tag{22}$$

The ratio of absorption for $\mathbf{E}\parallel\mathbf{B}$ to that for $\mathbf{E}\perp\mathbf{B}$ is of the order

$$(\hbar\Omega_{n'0}+\zeta)^{1/2}\zeta^{1/2}/\hbar\omega,$$

which is very small since normally $\zeta\ll\hbar\omega$.

EXPERIMENTAL RESULTS AND DISCUSSION

Measurements were made on samples having carrier concentrations of the order of 10^{16} cm^{-3}. It is known that carriers in InSb may freeze out on impurities under sufficiently high magnetic fields, depending on the carrier concentration and temperature. Hall measurements were made on the samples used in order to determine whether the phenomena observed were produced by free carriers. The measurements showed that the Hall coefficient at the temperature of optical measurement, ~13°K, had about the same value as at liquid-nitrogen temperature. The resistivity increased with increasing magnetic field, by a factor of 5 at the highest field of ~30 kG, while the Hall coefficient varied by only a few percent. Thus we conclude that the effect of free carriers was observed.

Transmission was measured at each frequency as the magnetic field was swept in the range 0 to $\gtrsim$30 kG. The two curves in Fig. 2 taken with $\mathbf{E}\perp\mathbf{B}$ radiation clearly show dips which correspond to peaks in absorption. The remaining curve illustrates the absence of absorption peaks with $\mathbf{E}\parallel\mathbf{B}$, in accordance with the theory of the resonance. In Fig. 3, the photon energies corresponding to the observed minima of transmission are plotted against the applied magnetic field, for a sample of 1.04×10^{16} cm^{-3} in carrier concentration. The solid curves converging to the energy $\hbar\omega=\hbar\omega_0=24.5\times10^{-3}$ eV at $B=0$ are drawn to fit the data points. These curves are consistent with (15) for the phonon cyclotron resonances. If the Landau levels are evenly spaced in energy, and if the spin splitting of each level is the same, then (15) can be written

$$\hbar\omega=\hbar\omega_0+(n'-n)\hbar\omega,$$

and $\hbar\omega$ would depend only on $\Delta n=n'-n$ irrespective of the sub-band n occupied by the carriers. Insofar as there is departure from the simple condition, carriers in different sub-bands will give resonances at somewhat different frequencies. This effect would lead to a smearing of the absorption peaks if the carriers are distributed over many sub-bands. With a carrier concentration of

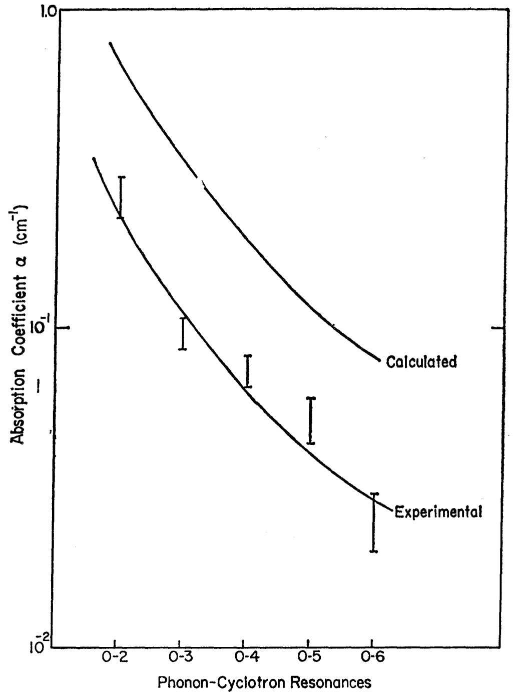

FIG. 4. Peak absorption coefficient of 0^+-n^+ resonances for $B=30$ kG.

1×10^{16} cm^{-3}, only the two lowest sub-bands, 0^+ and 0^-, of $n=0$ were occupied for $B>10$ kG and just one sub-band, 0^+, was occupied for $B>24$ kG. Above 20 kG, over 70% of the carriers were in the 0^+ sub-band. For our data, we can with good approximation consider only $\epsilon(n^+)-\epsilon(0^+)$. Calculations of Landau levels have been made by Palik and Wright[9] up to $n=4$ and by Pidgeon *et al.*[10] up to $n=5$. The curves up to $n=4$ are in good agreement with the calculations of the former authors, whereas the second calculation (which is considered to be more accurate) would give slightly lower curves, by $\sim3\times10^{-3}$ eV at the high fields. The 0^+-5^+ curve, however, agrees with the second calculation near 30 kG.

The absorption coefficient of the resonance peaks at 30 kG is plotted in Fig. 4. Also shown is a curve calculated from the theoretical expression (16a) with a coupling constant $\alpha=0.02$ and using the experimental resolution, $\sim3\times10^{-3}$ eV, for $\Delta\omega$. The measured values are lower by about a factor of 3 but they are reasonable in order of magnitude. Any broadening of the energy levels tends to reduce the resonance peaks. Such effect is not taken into account in the theory. The absorption merges with the background away from the resonance. Good estimates of the integrated absorption are not easily obtained. Figure 4 shows also that the way in which the measured absorption decreases with increasing Δn agrees satisfactorily with the variation shown by the theoretical curve. Finally, some measurements made on a sample of smaller carrier concentration ($n\sim0.75\times10^{15}$ cm^{-3}) showed that the resonance absorption is smaller by an order of magnitude in accordance with the theory. In this case, $\zeta=5\times10^{-5}\text{eV}\ll\hbar\Delta\omega\sim10^{-3}$ eV, and (16b) is the appropriate expression. We conclude that a clear understanding of the observed resonance phenomena has been obtained.

Figure 3 shows points lying below the 0-1 line which apparently do not represent phonon cyclotron resonances. The points appear to follow the dashed curves drawn through the origin which correspond to the harmonic cyclotron resonances 0^+-3^+ and 0^+-2^+, respectively.[3] However, neither set of peaks was detected with $E\parallel B$ radiation. According to Bell and Rogers,[6] the second harmonic of cyclotron resonance occurs with $E\parallel B$, whereas the third harmonic and the $\Delta n=2$ transitions with spin flip occur for $E\perp B$. Thus the points falling along the lower dashed curve but lying somewhat higher are more likely to be the 0^+-2^- combined resonance. This interpretation should be regarded as tentative. It should be pointed out that the points lying below the 0-1 solid curve represent absorption peaks which were among the strongest observed in the sample. On the other hand, none of these peaks was detected in the sample of $n=0.75\times10^{15}$cm^{-3}, which showed only the phonon cyclotron resonances. We should also point out that the Hall measurement showed no significant freeze-out of carriers in this sample as well. The variation in these absorption peaks is not yet explained.

It is seen in Fig. 2 that beside the dips given by the resonance absorption there is, with $\mathbf{E}\perp\mathbf{B}$, a general decrease of transmission with increasing magnetic field. The effect is more pronounced at lower photon energies, and it seems to be caused by the high-energy tail of cyclotron-resonance absorption. With $\mathbf{E}\parallel\mathbf{B}$, the transmission shows very little variation with the magnetic field, remaining nearly the same as at $B=0$. The peak absorption given by the phonon cyclotron resonance is small compared with the absorption at $B=0$. These observations indicate that there is a large part of the free-carrier absorption which is produced by indirect transitions not involving optical phonons. Presumably, these are indirect transitions involving acoustic phonons and transitions involving impurities.

[9] E. D. Palik and G. B. Wright, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic Press Inc., New York, 1967), Vol. 3, p. 442.

[10] C. R. Pidgeon, D. L. Mitchell, and R. N. Brown, Phys. Rev. **154**, 737 (1967).