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V-1. Magneto-Optical Phenomena

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§ 1. Introduction

Magneto-optical investigations in semiconductors have experienced a revolution during the last decade due to the advent of high magnetic fields, low temperatures, and pure single crystals. The present level of sophistication experimentally and theoretically has provided a powerful tool for quantitative analysis of the band structure of semiconductors and semimetals. New techniques and phenomena in magneto-absorption and magneto-reflection are now examining the relaxation and level broadening phenomena in magnetic fields. Interaction with the lattice, such as the polaron in a magnetic field, are observed by interband and intraband phenomena. Perhaps the most exciting future activities are the linear and nonlinear magneto-optical phenomena that are now possible with the availability of lasers in the near and far infrared. The latter opens up a whole new field of magnetic resonance. Theoretical treatment of classical nonlinear phenomena has been reported at Exeter. Today we now have a more general quantum treatment of nonlinear magneto-optical phenomena to all orders of electric field which considers a new class of nonlinear multiphoton resonances for interband transitions and intraband tunneling. Dispersion in the nonlinear regime to all orders is more general than that obtained by the perturbation treatment. Raman scattering and related effects in the presence of magnetic field are now possible. Nonlinear magneto-optics with the use of new and improved lasers and high magnetic fields is an unexplored area of research in semiconductors.

§ 2. Linear Magneto-Optics

Magneto-absorption in semiconductors, which was first observed in germanium[1] and indium antimonide,[2] has now been extended to other materials and to fields as high as 100,000 gauss or more. The data has been more thoroughly analyzed, fine structure has been identified, and band parameters determined in a self-consistent manner. In indium antimonide and indium arsenide,[3] the band theory included both the warping and nonparabolic terms in an 8×8 matrix to obtain quantitative comparison of theory and experiment, as shown in Fig. 1. The band parameters were determined in a computer program for best fit. The analysis is being extended to the inverted bands of mercury-telluride,[4] which has a band analogous to grey tin. The data in magneto-reflection yields two series; the allowed transition between the *s*-like valence band, the $p^{3/2}$ conduction band, and the forbidden transition between the two degenerate $p^{3/2}$ valence and conduction bands. Preliminary analysis determines the effective masses of the various bands from the experiment and the Kane ***k*-*p*** theory of the bands. The forbidden transition has now also been observed in tellurium.[5] Polarization with the electric field parallel or perpendicular to the *c*-axis permits two types of direct transitions to be observed in high magnetic fields.

Electric fields modulation in magneto-absorption and magneto-reflection has proved to be a very sensitive technique for extending the interband magneto-optical measurements. Vrehen[6] first used this to verify the predictions of Aronov[7] for the modification of the selection rules in cross-field magneto-absorption. Both allowed and forbidden transitions with $\Delta n=0$ and $\Delta n=\pm1$ are detected as shown in Fig. 2. The data is compared with the results of perturbation theory to second order in the electric field.[8]

In addition to improving the sensitivity, the electric field modulation sharpens the spectra lines since allowed and forbidden transitions yield negative and positive signals respectively. Electric field modulation has also been applied to magneto-reflection[9] by using the recently developed[10] piezo-reflectance and the electro-reflectance[11] techniques. These permit the observation of the oscillations up to the $p^{1/2}$ split-off band in germanium. These methods, including thermal modulation, should be useful in extending the magneto-reflection measurements to semimetals and metals.

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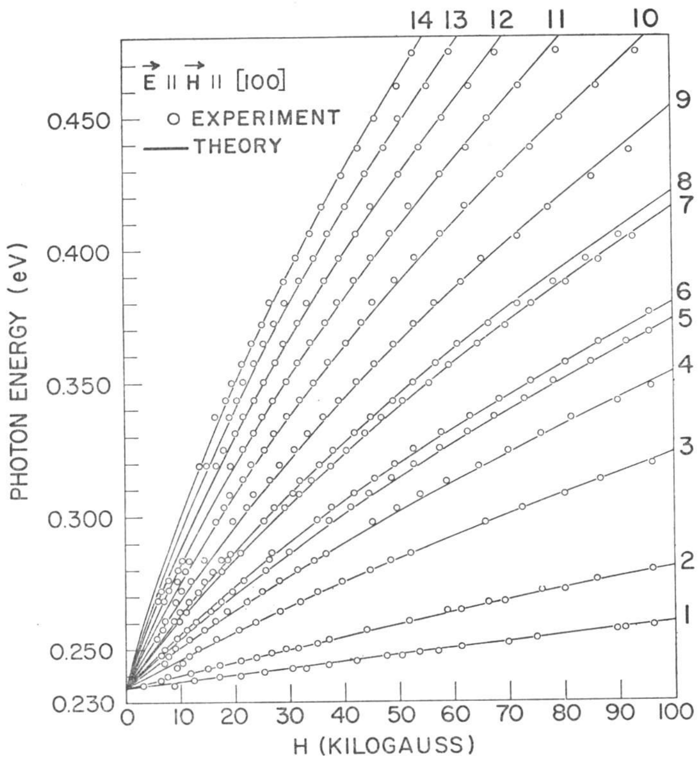

Fig. 1. Plot of principal transitions in InSb as a function of magnetic field for $E||H||[100]$. The solid lines represent the best theoretical fit to experimental data. The number next to each line indicates the quantum assignment. [After C. R. Pidgeon and R. N. Brown, ref. 3)].

§ 3. Photon Assisted Magneto-Tunneling

In order to explore the cross-field magneto-optical studies at high electric fields, both the theory and experimental techniques had to be modified. The experiments were performed on a back-biased germanium diode[12] in which photon energies below the energy gap were transmitted while both the electric and magnetic fields were adjusted. The theory has been treated in terms of nonparabolic bands to remove the limitation on electric field.[13] In the presence of high electric fields, photon assisted tunneling, better known as the Franz-Keldysh effect, is observed. With a magnetic field superimposed, the absorption tail below the energy gap is strongly effected, both by longitudinal and transverse magnetic fields.[14] This is readily understood by modifying the theory to include the magnetic field. The absorption coefficient then becomes

$$\alpha(H)=A\omega_c \sum \left[\frac{1}{(\varepsilon_n-\hbar\omega)^2} + \frac{3\beta}{2(\varepsilon_n-\hbar\omega)^{1/2}} e^{-\beta(\varepsilon_n-\hbar\omega)^{3/2}}\right], \quad (1)$$

where ω_c is the cyclotron frequency, A and β are coefficient given by the following expressions;

$$A=e^3\frac{E\mu}{2\hbar^3\omega c\eta m^2}|P_{cv}|^2\,; \quad \beta=\frac{4\sqrt{2\mu}}{3eE\hbar}\,;$$

where μ is the reduced mass, η the index of refraction, and P_{cv} the momentum matrix element.

In the case of the transverse magnetic field, the most suitable solution has been that obtained by using the nonparabolic representation of the energy bands for the tunneling problem. This gives a result in the quantum limit which is given by[15]

$$\frac{\alpha_\perp(H)}{\alpha_\parallel(H)}=e^{-\beta(\varepsilon_g-\hbar\omega)^{3/2}\,E/E_{\rm eff}}$$
$$\approx e^{-\beta(\varepsilon_g-\hbar\omega)^{3/2}}\varepsilon_g H^2/4m^*c^2E^2\,, \quad (2)$$

where $E_{\rm eff}=(E^2-\varepsilon_g H^2/2m^*c^2)^{1/2}$, deduced from a Lorentz transformation. Thus the slope of the transverse magneto-tunneling is reduced quadratically as the magnetic field is increased. The diode experiments have also shown oscillations

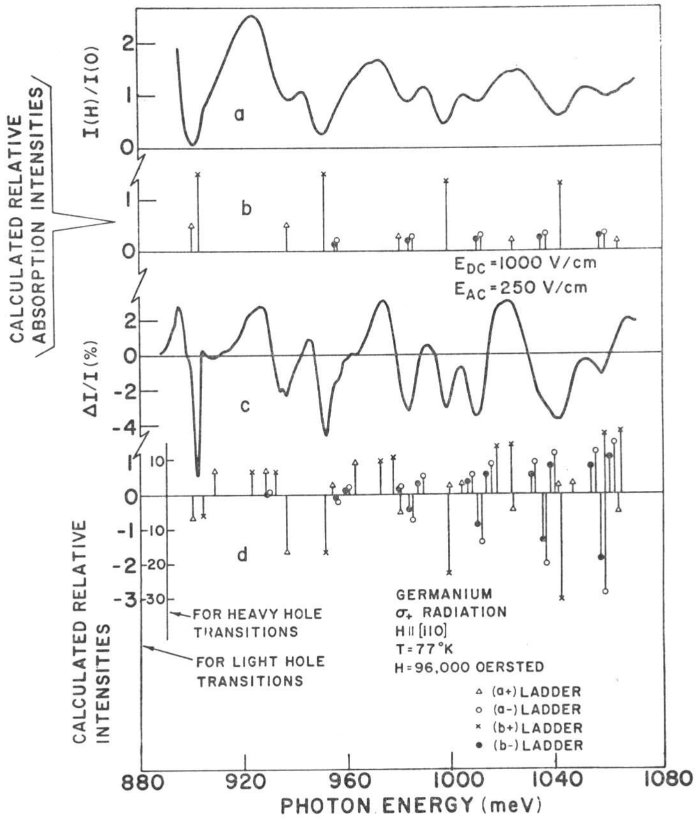

Fig. 2. Magneto-absorption and cross-field differential absorption in germanium: (a) Magneto-absorption spectrum; (b) calculated spectrum; (c) cross-field spectrum; (d) calculated differential spectrum. Positive lines are forbidden transitions, and negative lines are allowed. [After Q. H. F. Vrehen, ref. 8)].

in the absorption tail.[16] These are due to the energy term in the eigenvalue which reduces the energy gap

$$\Delta\varepsilon_g = -\frac{c^2E^2}{2H^2}(m_1+m_2)\,, \qquad (3)$$

where m_1 and m_2 are the effective masses of the conduction and valence bands respectively. Indeed by looking at the first minimum below the gap, Vrehen[17] has shown that within the appropriate limit for light hole electron transitions, this expression holds and the shift of the energy gap plotted as function of E^2/H^2 fits a straight line quite well. To see additional minima it was necessary to look at the polarization effects[18] below the energy gap both in the Faraday and Voigt configurations which enhanced the oscillations as shown in Fig. 3. This permits a quantitative study of the effect of the electric field on the lower quantum levels of the valence band. The theory as obtained from the perturbation treatment in germanium gives a good semi-quantitative agreement with the experimental results. However, absolute

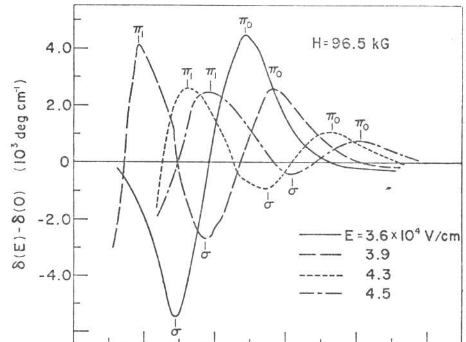

Fig. 3. Photon assisted magneto-tunneling below the energy gap in crossed fields. The oscillations are observed by polarized light in germanium diodes for low quantum values of the holes. [After M. Reine, W. Zawadzki, and Q. H. F. Vrehen, ref. 14)].

magnitudes of spacing were not quite correct. Nevertheless, the variation of the motion of levels with increased electric field were correctly predicted.

According to Zak and Zawadzki[19] the effective mass approximation for a Bloch electron in crossed electric and magnetic fields holds only if $eEa > \hbar\omega_c$. However, even within this limit the Aronov approximation breaks down, and the two-band Hamiltonian (interaction of s-band and three degenerate p-bands) yields the following equation which is more appropriate for cross fields.[20]

$$\left[\frac{p_y^2}{2m^*} - \alpha y + \frac{m^*}{2}\left(\omega_c^2 - \frac{2e^2E^2}{m^*\varepsilon_g}\right)y^2 + \frac{3}{8}\frac{\hbar^2}{m^*}\{y - (\varepsilon_g+\varepsilon)eE\}^{-2}\right]\varphi(y) = \lambda\varphi(y), \quad (4)$$

where $\alpha = \hbar\omega_c k_x - eE(\varepsilon_g + 2\varepsilon)/\varepsilon_g$
and $\lambda = \varepsilon(\varepsilon+\varepsilon_g)/\varepsilon_g - \hbar^2k_x^2/2m^* - \hbar^2k_z^2/2m^*$.

The solution distinguishes between electric field and magnetic field dominated phenomena, the critical field being given by

$$E = H(\varepsilon_g/2m^*c^2)^{1/2}. \quad (5)$$

The nonparabolic Hamiltonian also predicts higher order transitions $\Delta n = \pm 3$ and even second harmonics with DC electric field $\Delta n = \pm 2$ for intraband cyclotron resonance.[21] An effective "spin orbit term" modifies the anomalous g-factor as well.

§ 4. Line and Level Broadening

It has been known from quantum transport theory[22] that the scattering of electrons is modified by high magnetic fields.[23] Gurevich and Firsov[24] had similarly predicted oscillatory effects in the magneto-resistance in semiconductors when the optical phonon frequency is a multiple of the cyclotron frequency. However, very little theoretical work has been done with the analogous problem involving photons in magnetic fields. Shin[25] has treated scattering and level broadening by density matrix formalism both for intraband and interband transitions in a general way, thereby extending the Kubo formalism[26] to this problem. In another context we have considered the consequences of Kubo's Landau level broadening to explain the magnetic field dependence of the threshold current of a magneto-optical laser.[27] The significance of both of these results is that for the first time there exists a connection between formal theory of level broadening and magneto-optical experiments. Meyer[28] showed that by applying the "golden rule" to second order, that the cyclotron resonance linewidth varies directly as the magnetic field. Shin's treatment extends this result and also Kubo's concept of Landau level broadening to consider linewidth as a function of frequency and quantum number as well as magnetic field. His results account quantitatively for the increased damping of the oscillations in the interband and intraband magneto-optical experiments with frequency and with quantum number, At the same time, optical phonon scattering resonances are predicted for both. This is the optical analogue of the Gurevich-Firsov effect. Some of the consequences of this theory have not been examined experimentally. Related theoretical treatment of interband line shapes for magneto-optical absorption has been considered by Korovin and Kharitonov.[29]

Kubo's concept of Landau level broadening in high magnetic fields, in which the singularity of the density of states is removed, is tested by the threshold measurements of the magneto-optical laser. According to Kubo the density of states varies as $H^{2/3}$ and therefore the threshold for the laser would vary as $H^{-4/3}$. Indeed when this theoretical slope is compared to the experimental data for the lowest transition in indium antimonide,[30] the best fit is obtained as shown in Fig. 4. Thus it appears that for the

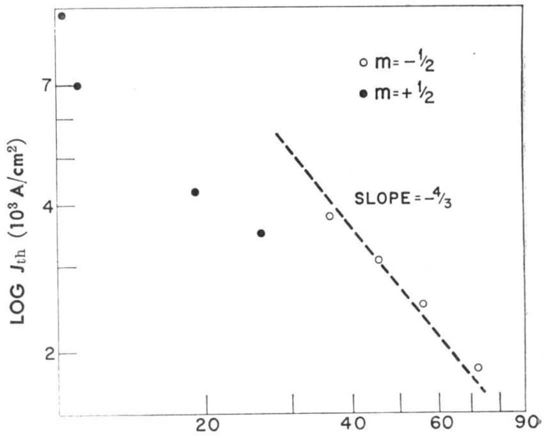

Fig. 4. Threshold current for an InSb diode laser as a function of magnetic field. The dotted line is theoretical curve for $J_t \sim H^{-4/3}$. The open circles are data for transitions between lowest conduction band level and valence band. [After B. Sacks and B. Lax, ref. 29)].

first time we have a means of experimentally investigating the fundamental behavior of the density of states in a magnetic field.

§ 5. Cyclotron Resonance with Lasers

For sometime now it has been apparent that with large fields of the order of 100 to 200 KG it should be possible to do resonance experiments in the far infrared region of the spectrum if a suitable monochromatic source were available. The development of the cyanide laser[31] which operates at 337 microns appeared to be a suitable instrument for this purpose. In order to demonstrate the feasibility of the technique we have decided to use such a spectrometer[32] to look at cyclotron resonance in *p*-type germanium. At room temperature resonance of the light hole was barely perceptable. However, at 165°K the light and heavy hole were easily distinguished and several quantum transitions appeared. Additional structure was resolved and different quantum transitions could be selectively enhanced by varying the temperature between 20°K and 50°K, or by varying the laser power, as shown in Fig. 5. With this laser source, which has peak powers of the order of 1 watt, the intrinsic linewidth was broadened by the laser. When low power CW laser becomes available, the intrinsic linewidth could be measured. On pulsed operation with 10 watts peak, it should then be possible to study the well-known nonlinear phenomena of resonance in the submillimeter range in which population saturation of levels are possible and parametric interactions are induced. Thus the 300 micron laser can become a source for pumping 3-level lasers involving both the electric dipole and magnetic dipole transitions to provide tunable sources of radiation in the submillimeter region.

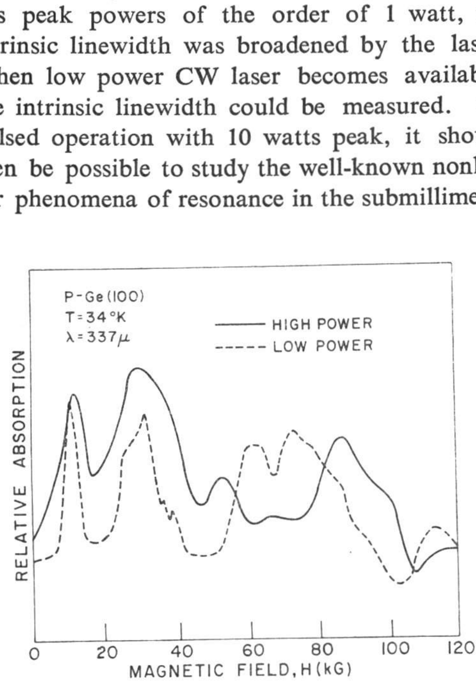

Fig. 5. Cyclotron resonance absorption in *p*-type germanium with far infrared cyanide laser. Light and heavy hole resonances at extreme left and right respectively. Lines inbetween are transitions for low quantum numbers. [After Button, Lax, and Gebbie, ref. 32)].

§ 6. Nonlinear Magneto-Optics

Four years ago we had pointed out the possibilities of intraband nonlinear magneto-optical phenomena in semiconductors,[33] if and when suitable lasers of photon energies below the energy gap were available. With the advent of the CO_2 laser, this now has been realized, and in addition to the intraband magneto-plasma experiments, interband experiments with magnetic fields have now begun.[34] A formal quantum treatment which extends the classical and perturbation treatments has also been developed to consider these particular problems. The semi-classical treatment considers free carriers with the appropriate effective mass or bound electrons distributed over an energy band with the appropriate oscillator strength in a nonlinear response to a strong oscillating electric field. The various mechanisms for inducing nonlinearity include: (1) higher order nonlinear restoring forces of the bound electron; (2) the $\boldsymbol{v} \times \boldsymbol{h}$ term in which the laser induced RF magnetic field is sufficiently large to generate harmonics. Other nonlinear mechanisms in the intraband case include (3) the variation of the scattering time τ with energy, (4) the nonparabolic equations of motion which contain higher order terms in momentum when expanded. Each of these treatments is essentially a perturbation or reiterative approximation in which the equations of motion are expanded as a power series in the electric field to second or third order. We have solved the analogous problem quantum mechanically by carrying out time dependent perturbation solution to second order for interband transitions.[35] This has the virtue, à la Braunstein[36] of considering the importance of higher bands and relevant selection rules for allowed and forbidden transitions in the presence of a magnetic field for two photon or even three photon processes. The perturbation results are important in that they provide additional information about higher bands which supplement the single photon or linear magneto-optical studies. However, there is a limitation to the perturbation treatment since it is only admissible in a limited range of electric field which exceeds a threshold that bridges the gap between the linearly and highly nonlinear regime. In order to treat the problem properly we have taken the tunneling formalism analogous to that of Keldysh[32] and have applied it to the magneto-optical problem. It was necessary to develop a set of exact eigen-functions in the presence of both the oscillating electric field and the DC magnetic field in which the wave function contains the electric field to all orders. For the case where the electric and magnetic field are parallel to one another, the problem becomes a simple extension of the Keldysh solution. Nevertheless in evaluating the matrix elements for the transition probabilities it becomes apparent that a nonlinear multi-photon resonance effect can be observed. The condition for this resonance is given by

$$l\hbar\omega=\varepsilon_g+\left(n+\frac{1}{2}\right)\hbar\omega_c+\frac{e^2E^2}{em\omega^2}\cdot \quad (E\|H) \quad (6)$$

Keldysh considered the interband case and the above equation is the magneto-optical analog of his particular problem. Its importance is that it predicts a multi-photon resonance to higher orders by virtue of the high electric field. The matrix elements for these transitions then can be studied as a function of both electric and magnetic fields.

From the theoretical point of view, perhaps the most interesting interband nonlinear problem in semiconductors is the transverse case in which the exact solution for the time dependent wave function becomes more complicated and therefore its solutions correspondingly richer. The wave function for the time dependent equation for oscillating electric field transverse to the magnetic field takes the form

$$\Psi(x,t)\sim e^{ix\alpha(t)}\Phi_n(x-\beta(t))e^{-i(n+1/2)\omega_c t}, \quad (7)$$

where $\hbar\alpha(t)$ is the classical time dependent momentum, $\beta(t)$ the conjugate oscillating displacement of the magnetic orbit. In this case the resonance condition is altered when the matrix elements are calculated in that

$$l\hbar\omega=\varepsilon_g+\left(n+\frac{1}{2}\right)\hbar\omega_{c1}+\left(n^1+\frac{1}{2}\right)\hbar\omega_{c2}+\frac{e^2E^2}{4m_1(\omega^2-\omega_{c1}^2)}+\frac{e^2E^2}{4m_2(\omega^2-\omega_{c2}^2)}\cdot \quad (E\perp H) \quad (8)$$

The resonance phenomenon here is analogous to those for the longitudinal case where the electric and magnetic field are parallel with two exceptions. First of all, the selection rules for the interband transitions, *i.e.*, $\Delta n=0$ are relaxed and all transitions are possible, namely, $\Delta n=\pm1$, $\Delta n=\pm2$, etc. The origin of this breakdown of selection rules is exactly analogous to that obtained by Aronov for his cross-field problem in the DC electric field. The second set of terms in eq. (8) are the high frequency shift of the energy gap analogous to that with crossed electric fields. In this case, however, this modulation of the energy can become positive or negative, depending on whether the frequency of the laser light is higher or smaller than the cyclotron frequency of the holes and the electrons. In the limit of zero frequency, this reduces to Aronov's expression of eq. (1). From the quantum point of view this energy term exists because the center of the Landau orbit oscillates at the characteristic frequency associated with the laser. Both interband and intraband resonances can be simultaneously observed. In the intraband transitions, a tunneling phenomenon between magnetic and sub-bands also occurs because of the large resonant energy obtained by the holes and electrons.

On the experimental side it appears that the most suitable candidates for these experiments in the immediate future are those in which low gap materials such as lead telluride, lead selenide, indium antimonide, indium arsenide are involved, particularly for the use with the CO_2 laser. In the nonlinear magneto-absorption it is now possible to pump a laser by this multiphoton process as demonstrated for the non-magnetic case by Patel and co-workers.[38] We can now, however, enhance the pumping efficiency by tuning

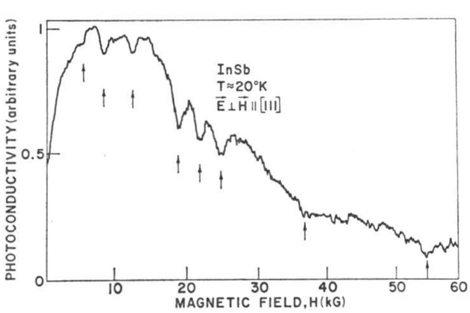

Fig. 6. Nonlinear multiphoton interband resonance in InSb. The minima in the photoconductivity correspond to the absorption of three-photons from the 10.6 micron emission of the CO_2 laser. [After Button, Lax, Weiler, and Reine, ref. 34)].

the magnetic field to either two or three photon resonances. Such interband multiphoton magneto-optical resonances in InSb and PbTe have now been observed. The appearance of these resonances as shown in Fig. 6 can be correlated with two photon absorption by means of eq. (8) where the intraband energy terms are small. Furthermore, this now opens up the possibility of providing an efficient pump for a four-level cyclotron resonance laser in which the two levels $n=1$ in both the valence and conduction band are saturated and simultaneously the intraband laser between $n=0$ for which the transition probability is high provides a sink which depeletes the lower layer and thereby inverting the interband levels $n=1$ and $n=0$ both in the valence and conduction bands. This then has the possibility of simultaneously operating three lasers at once, namely the interband and two intraband at the electron and hole cyclotron frequency respectively. Even if the latter are not adequately inverted to produce laser action sufficient spontaneous emission may occur to provide a useable course in the infrared or millimeter region which could occur from 10 to 100 microns in such materials as indium antimonide and lead telluride.

§ 7. Magneto-Raman Effects

The electromagnetic field can probe the interaction of the electron with the collective or many-body excitations of a solid. The addition of a magnetic field profoundly affects the motion of the electron and modifies the spectral behavior of the interaction. This has been clearly illustrated by the magnetic effect of the polaron[39] in the interband spectrum in InSb. Another experiment that would display a magnetic effect is the Raman scattering in which the optical phonons are coupled to the magnetoplasma. The effect of $H=0$ has been observed by Mooradian and Wright.[40] The magnetic field dependence would measure the coupling between the phonons and the plasma. In its simplest form the plasma frequency is given by:

$$n^2=\varepsilon_\infty\left[1-\frac{\Omega_p{}^2}{\omega^2-\omega_t^2}-\frac{\omega_p{}^2}{\omega(\omega\pm\omega_c)}\right]=0,$$

where $\Omega_p{}^2=(\varepsilon_0-\varepsilon_\infty)/\omega_t{}^2$, ω_t is the transverse optical phonon frequency and ω_p the electron plasma frequency $\omega_p{}^2=Ne^2/m^*\varepsilon_\infty$.

Theoretical treatments of scattering of light with and without magnetic fields has been considered. Platzman[41] and McWhorter[42] treated elastic and inelastic scattering of plasmons. Their treatment has been extended by Wolff[43] to take into account the effect of a magnetic field. Cross section for nonparabolic bands were obtained. The result shows that substantial enhancement occurs when the light frequency corresponds to cyclotron resonance and its energy is near the energy gap of the semiconductor. Thus low-gap materials appear ideal for such experiments.

§ 8. Conclusion

It appears that the recently developed techniques of piezo, electro and thermal reflectance techniques should enhance the usefulness of the linear magneto-optical measurements in semiconductors and semimetals. The use of low power lasers in the infrared will advance resonance and high resolution spectroscopy and high power lasers now make possible the investigation of new nonlinear magneto-optical phenomena and elastic and inelastic scattering of light beams by magnetoplasma in solids.

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DISCUSSION

Burstein, E.: 1) Raman scattering by plasmons in crystals lacking a center of symmetry arises both from the coupling of the plasmons with longitudinal phonons, as pointed out by Mooradian and Wright, and from the electro-optic effect of the macroscopic "longitudinal" electric field (Burstein and Iwasa, to be published) which Mooradian and Wright did not consider. These mechanisms do not play a role in crystals with center of symmetry. Can you say anything about the magnitude of the scattering efficiency by electrons or plasmons in a magnetic field for crystals with inversion symmetry?

2) Whether 2 photon or 3 photon processes occur is determined by the parity of the initial and the final states. Is this the case for the two photon processes in PbTe which you attribute to transitions at the L point?

Lax, B.: We have no theory for the cross-section of the magneto-raman scattering where the frequency shift is by the magneto-plasmon-phonon frequency, Wolff's analysis does not apply since he considered a single electron interaction.

Since in PbTe the two photon process occurs fairly high in the band the theory has not been worked out in detail.

Cardona, M.: InSb while cubic, has no inversion symmetry. Hence two photon transitions between Γ_{15} and Γ_1 are allowed, with the Γ_{15} conduction band as an intermediate state. However since the inversion symmetry is only weakly violated, the two photon transitions are only weakly allowed. In PbTe there is parity at the L point and hence since the direct gap is parity-allowed for one photon, it should be forbidden for two photons.

Lax, B.: The transitions take place fairly high in the band even with two photons.

Hasegawa, H.: How essential or not essential is the nonparabolicity of bands in the predicted multi-photon processes between Landau levels?

Lax, B.: The selection rules are not affected by this, since the momentum operator is on the Bloch functions, not on the envelope functions, in the perturbation treatment. The latter give $\Delta M=0$ from the orthonormalization of the harmonic oscillator functions.

Pikus, G. E.: As reported in our paper, Dr. Aronov and I have also investigated theoretically the light absorption in crossed electric and magnetic fields. I would like to point out some differences between our result and those reported by Prof. Lax. According to the expression quoted by Prof. Lax, the *H*-square terms in the exponent is proportional to $(E_g-\hbar\omega)^{3/2}$ whereas we get $(E_g-\hbar\omega)^{5/2}$. According to our result, therefore, in the region near the absorption edge, the effect of the magnetic field should be weaker than that given by the expression of Prof. Lax.

Lax, B.: In contrast to your approach, we derived our result with the use of the WKB approximation. At present there is no clear-cut answer as to which of the two expressions is correct.