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V-7. The Effects of Band-Population on Interband Magneto-Optical Phenomena

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The magneto-conductivity tensor is calculated for an isotropic semiconductor with partially occupied bands. Large changes in the interband contributions to the high frequency Hall conductivity are generated by the band-population. These are associated with the Burstein-Moss optical band edges in a magnetic field. The de Haas-van Alphen oscillatory and the non-oscillatory contributions to the interband Faraday rotation and to the magneto-plasma edge are calculated and compared with experiment. The oscillatory Faraday rotation and the non-oscillatory magneto-plasma edges were measured in n-type PbS and the results are reported here.

§ 1. Introduction

The addition of free carriers to an empty band of a solid significantly alters the interband contributions to the magneto-conductivity. These terms have dispersive as well as absorptive character so that the magneto-phenomena over a wide frequency range are affected. The primary effect has been observed experimentally by measurement of the frequency and temperature dependent Faraday rotation in n-type PbS.[1] A detailed theoretical fit was obtained using known band parameters.

A physical description of the band-population contributions to magneto-optical effects has been given[1] in terms of the absorptive and dispersive consequences of the Burstein-Moss[2] effect for left- and right-circularly-polarized radiation (*lcp* and *rcp*). Stern[3] suggested another possible physical interpretation in terms of the spin-dependent polarization of the free carriers. The first description directly relates the new effects to interband terms in the conductivity tensor which can be experimentally measured while the spin dependent dielectric polarization is not directly observable.

Several new band-population effects are reported here together with a more complete theory which includes the de Haas-van Alphen (DHVA) type oscillatory terms previously neglected. We have experimentally observed the DHVA terms in the interband Faraday rotation below the band gap in n-type PbS and report the results. The free-carrier magneto-plasma edge is also shown to contain DHVA terms which have possibly been observed[4] but were attributed to another physical mechanism. The non-oscillatory magneto-plasma effect likewise is affected by the band population as we have also observed in n-type PbS.

§ 2. Faraday Rotation

A. Conductivity formulation

The Faraday rotation per unit length $\theta(\omega)$, for cubic crystals in low absorbing frequency ranges, is given by[5,6]

$$n(\omega)\theta(\omega)=\frac{2\pi}{c}\sigma_{xy}^{(1)}(\omega)\ , \tag{2.1}$$

where $n(\omega)$ is the frequency dependent index of refraction in zero field and $\sigma_{xy}^{(1)}(\omega)$, the frequency dependent Hall conductivity, is the real part of the off-diagonal component of the conductivity tensor. The magnetic field is in the $+z$ direction.

The expression for the frequency dependent Hall conductivity obtained by using semiclassical perturbation theory is given by[5]

$$\sigma_{xy}^{(1)}(\omega)=\frac{Me^2}{\hbar m_0{}^2 V} \times \sum_{i<f}\left\{\frac{|<f|\pi^-|i>|^2}{\omega_{fi}^2-\omega^2}-\frac{|<f|\pi^+|i>|^2}{\omega_{fi}^2-\omega^2}\right\}F_{if}\ , \tag{2.2}$$

where M is the number of equivalent bands (four for PbS) and where $\pi^{\pm}=(\pi_x\pm i\pi_y)/\sqrt{2}$ is the kinetic momentum operator.[8] We have written the sum over states so that the initial states are restricted to lie at lower energy than the final states ($\hbar\omega_f-\hbar\omega_i=\hbar\omega_{fi}>0$). The difference in occupation between the initial and final states is given by the Fermi function $F_{if}=F_i-F_f$.

We restrict the calculation to populated bands which are spherical and non-degenerate and are in the parabolic approximation. This approximation is well satisfied for n- and p-type PbS for which a theoretical spherical approximation has been developed for band extrema at the L-point of the Brillouin zone.[7] We also include the contributions to the valence and conduction band parameters from other deep-lying bands since major DHVA terms vanish for the two band model. We necessarily include the spin and spin-orbit mixing of bands since otherwise the effects vanish. An earlier calculation of the band-population contributions to the magneto-conductivity[9] neglected the spin and thus missed the major terms of interest.

The intraband (cyclotron resonance) terms of eq. (2.2) are evaluated using the selection rule[8] $\langle f\,|\,\pi^+\,|\,i\rangle = \hbar^2(m_0{}^2/m_c{}^2)\sqrt{(n+1)s}\,\delta_{n'-1,n}\delta(k_z{}'-k_z)$ where the conduction band is specified as the partially occupied band and $s=eB/\hbar c$. Other quantum numbers which play no role are neglected. The result for the free carrier case (*FC*)

$$(FC)\quad \sigma_{xy}^{(1)}(\omega)=\frac{-Me^2}{\hbar s}\,\frac{\omega_c{}^2}{(\omega_c{}^2-\omega^2)}\,\frac{(N_\alpha+N_\beta)}{V}\,, \tag{2.3}$$

gives the usual expression for the free carrier Faraday rotation when used in eq. (2.1). The total number of free carriers ($N=N_\alpha+N_\beta$) in the Kramers' sub-bands α and β are constant for each of the equivalent extrema so that no DHVA type terms appear in this approximation.

The eigenvalues for the energy levels in the presence of magnetic field are given by

$$\varepsilon_j(n,\,k_z)=\varepsilon_j(0)+(n+\tfrac{1}{2})\hbar\omega_j+\hbar^2k_z{}^2/2m_j\pm g_j\beta H/2\,, \tag{2.4}$$

where j is the band index and n is the Landau quantum number. The effective mass m_j, g-factor g_j and cyclotron frequency $\omega_j=eB/m_jc$ are normally positive for conduction bands and negative for valence bands. The selection rules for the interband transitions are given by $\langle f|\pi^\pm|i\rangle=\pi^0_{j',j}\delta_{n'n}\delta_{\sigma'\mp1,\sigma}\delta(k_z{}'-k_z)$ for spherical bands in the parabolic approximation where σ is the z component of the total angular momentum which transforms like the states at a given band in the absence of a magnetic field. Similarly $\pi^0_{j',j}$ is evaluated at the band edge in zero field. The magnetic field dependent corrections to $\pi^\pm_{cv}$ are negligible for the cases we consider (*i.e.* large g-factors).[10]

With this selection rule, the interband (*IB*) contributions to eq. (2.2) are given by

$$(IB)\quad \sigma_{xy}^{(1)}(\omega)=\frac{M\hbar e^2|\pi^0_{cv}|^2}{m_0{}^2V}\times\sum_{n,\varepsilon}\left\{\frac{F_+{}^\alpha-F_-{}^\beta}{[\varepsilon^2-(\hbar\omega)^2]}-\frac{[2(g_c+g_v)\beta B\varepsilon](F^0-1)}{[\varepsilon^2-(\hbar\omega)^2]^2}\right\}, \tag{2.5}$$

where the sum over k_z is replaced by a sum over the energy by relating the conduction band energy scale eq. (2.4) to the interband energy scale

$$\varepsilon^\pm_{cv}=\varepsilon^0_{cv}+(n+\tfrac{1}{2})\hbar(\omega_c-\omega_v)+(\hbar^2k_z{}^2/2)(m_c{}^{-1}-m_v{}^{-1})\pm(g_c+g_v)\beta H/2\,. \tag{2.6}$$

In eq. (2.5), ε is given by eq. (2.6) omitting the last term and the approximation is made $g_c\beta B\ll E_F{}^0$. The Fermi occupation functions $F_\pm$ are functions of the variable $[(\varepsilon-\varepsilon^0_{cv}-E'_{F\pm})/kT']$ where $E'_{F\pm}=[(1-m_c/m_v)E_F{}^0\pm(g_v+(m_c/m_v)g_c)\beta B/2]$ and $T'=(1-m_c/m_v)T$ and $\varepsilon_F{}^0$ is the Fermi level in zero field measured relative to the conduction band edge. The first term of eq. (2.5) gives contributions to the conductivity only in the vicinity of the Burstein-Moss energy $\varepsilon^{0'}_{cv}=\varepsilon^0_{cv}+(1-m_c/m_v)\varepsilon_F{}^0$. The second term gives the contributions to the conductivity which correspond to the usual interband conductivity reduced by the band population blockage.

The major DHVA oscillatory terms occur in the first term of eq. (2.5) which we evaluate in a form which emphasizes their physical origin. The approximation is made $kT\ll E_F$.

$$(IB)\quad \sigma_{xy}^{(1)}(\omega)=\frac{M\hbar e^2|\pi^0_{cv}|^2}{m_0{}^2[(\varepsilon^{0'}_{cv})^2-(\hbar\omega)^2]}\,\frac{N_\alpha-N_\beta}{V}+II\,. \tag{2.7}$$

The effects we are considering thus depend directly on the difference in population between the two Kramers' states of the populated band. This is not required to remain constant. The evaluation of eq. (2.7) is straightforward.[11]

$$(IB)\quad \sigma_{xy}^{(1)}(\omega)=\frac{\sqrt{2}M}{\pi^2}\,\frac{e^2(\mu_{cv})^{3/2}|\pi^0_{cv}|^2}{\hbar^2m_0{}^2[(\varepsilon^{0'}_{cv})^2-(\hbar\omega)^2]}\left\{(\varepsilon_F{}^{0\prime})^{1/2}(g_v+(m_c/m_v)g_c)\beta B+\frac{\pi(\beta B)^{1/2}m_ckT}{m_0(\mu_{cv})^{3/2}}\sum_{r=1}^{\infty}\frac{(-1)^{r+1}\sin[r\pi(g_v+(m_c/m_v)g_c)\mu_{cv}]\sin(2\pi rE_F{}^0/\hbar\omega_c+\pi/4)}{r^{1/2}\sinh 2\pi^2rkT/\hbar\omega_c}\right\}+II\,, \tag{2.8}$$

where $(\mu_{cv})^{-1}=(m_c/m_0)^{-1}-(m_v/m_0)^{-1}$ and where M is the number of equivalent bands. It should be noted that the effective mass and g-factors are those obtained by summing over all bands and that the valence band parameters are normally negative.

B. Experimental

The non-oscillatory part of eq. (2.8) is that obtained earlier in different form[1] and exhibits the low frequency tail discussed previously. The oscillatory part of eq. (2.8) is observable in the Faraday rotation measured below the energy gap. The experimental data for two n-type PbS samples at $T\approx 40°$K are plotted in Fig. 1 for frequencies below the gap. The period of the oscillations in Fig. 1 is frequency independent, below the gap, but is concentration dependent. Since only a single period was observed for each sample at this temperature, the maxima, minima and crossover points were plotted vs. $1/B$. A linear relationship was observed. The carrier concentrations were calculated from the periods (assuming four equivalent band extrema) and the values agreed with those obtained from Hall measurements within 20%.

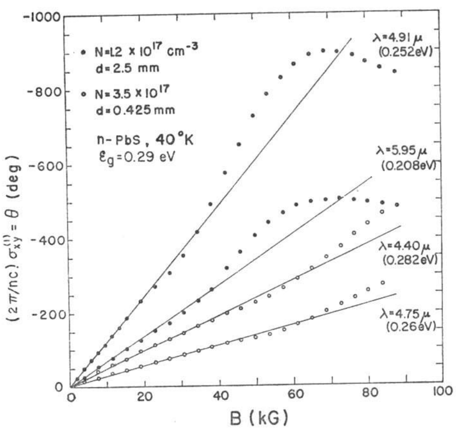

Fig. 1. The Faraday rotation measured in n-type PbS at 40°K is plotted for selected frequencies below the band gap. The period of the oscillatory part is frequency independent over a range twice that shown .

§ 3. Magneto-Plasma

The magneto-plasma edges for $lcp(+)$ and $rcp(-)$ radiation are determined by the zeros in the real part of the dielectric constant $\varepsilon_{\pm}{}^{(1)}(\omega)=[1-(4\pi/\omega)\sigma_{\pm}{}^{(2)}(\omega)]$ where $\sigma_{\pm}{}^{(2)}=\sigma_{xx}^{(2)}\pm i\sigma_{xy}^{(1)}$. The intraband and interband population contributions to $\sigma_{xx}^{(2)}$ have been treated in detail[6,9] but neglecting possible spin dependent population effects. We repeat the calculation for $\sigma_{xx}^{(2)}$ but include the spin and spin-orbit interaction. The details are similar to those for the calculation of $\sigma_{xy}^{(1)}$ and will not be repeated. The results are given by

$$\sigma_{xx}^{(2)}(\omega)=-\frac{Me^2\omega}{m_0{}^2V}\left\{\frac{m_0{}^2(N_\alpha+N_\beta)}{m_c(\omega_c{}^2-\omega^2)}\right.$$
$$\left.-\left[\sum_{n,\varepsilon}\frac{|\pi_{cv}^0|^2}{\varepsilon[\varepsilon^2-(\hbar\omega)^2]}\right]+\frac{|\pi_{cv}^0|^2(N_\alpha+N_\beta)}{(\varepsilon_{cv}^{0\prime})[(\varepsilon_{cv}^{0\prime})^2-(\hbar\omega)^2]}\right\}, \tag{2.9}$$

where the first is the dispersive part due to intraband transitions, the second is the dispersive part due to interband transitions of the pure crystal and the third part is the decrease in the dispersive part due to the population blockage of transitions below the Burstein-Moss edge. It is worth noting that no DHVA terms appear in the expression and in fact, to first order, the expression is the same as for zero magnetic field.

The plasma edges are obtained from solutions to the equation

$$\frac{\Omega_p{}^2(\omega_c\pm\omega)}{\omega(\omega_c{}^2-\omega^2)}=\varepsilon_{\pm}^{(1)}(\omega)\,, \tag{2.10}$$

where $\varepsilon_{\pm}^{(1)}(\omega)=[\varepsilon^{(1)}(\omega)]_{IB}\pm(4\pi/\omega)[\sigma_{xy}^{(1)}(\omega)]_{IB}$ and $\Omega_p{}^2=4\pi Ne^2/m_c$. The term on the right of eq. (2.10) is usually replaced by the frequency dependent dielectric constant for the pure crystal. Dresselhaus and Dresselhaus[9] discuss the shifts of the free carrier plasma edges due to band-population contributions to $\sigma_{xx}^{(2)}$ but in most cases these are small. The band-population contributions to the plasma edge shifts through $\sigma_{xy}^{(1)}$ are large and, in addition, contain large DHVA type terms. Thus the proper high frequency dielectric constant is not the same for $l$cp and rcp as is commonly assumed. If the plasma edges are well below the interband edges then the terms on the right $[\varepsilon^{(1)}(\omega)]_{IB}$ and $[\sigma_{xy}^{(1)}(\omega)]_{IB}$, are essentially constant over the frequency ranges $\omega\ll\omega_l$ and $\omega_l\ll\omega\ll\omega_{cv}$ where ω_l is the reststrahl frequency. The plasma edges including the DHVA terms can be calculated using the appropriate dielectric constant ε_0 or ε_∞ and the low frequency value for $[\sigma_{xy}^{(1)}]_{IB}$ obtained as in § 2. It is worth noting that in polar crystals, where $\varepsilon_\infty\ll\varepsilon_0$, there are free carrier plasma edges above and below the reststrahl frequency for a range of carrier concentrations.

The plasma edges $\omega_\pm$ obtained from eq. (2.10) are given by

$$\omega_{p\pm}=\omega_p\pm\left[2\pi(\omega_p/\Omega_p)^2\sigma_{xy}^{(1)}(\omega)+\frac{\omega_c}{2}\right], \quad (2.11)$$

where $\omega_p{}^2=\Omega_p{}^2[\varepsilon^{(1)}(\omega)]_{IB}^{-1}$. In solving eq. (2.10) we have adopted the approximation $[4\pi\sigma_{xy}^{(1)}/\omega]_{IB} \ll [\varepsilon^{(1)}(\omega)]_{IB}$.

A. Experiment

Equation (2.11) predicts a departure from the value ω_c for the separation of the *lcp* and *rcp* plasma edges. It appears as if the interband correction term might either increase or decrease the spacing. If both valence and conduction band transform like states of total angular momentum $J_{1/2}$ as is the case for PbS, then the dominant contribution to the *g*-factor is positive for the conduction band and negative for the valence band.[7] In this case, the spacing is decreased and goes to zero in the limit of large spin-orbit interaction.

We plot in Fig. 2 the measured plasma edge splitting for *n*-type PbS at 80°K together with the splittings calculated without and with the interband correction term. Good agreement is obtained for the latter where the value for the correction was obtained from other experiments.[1] It is stressed that this correction *must* be applied for materials with large spin-orbit interaction in order to obtain the correct value for the effective mass.

It is possible that the DHVA-type oscillatory terms of $\sigma_{xy}^{(1)}$ have been observed experimentally

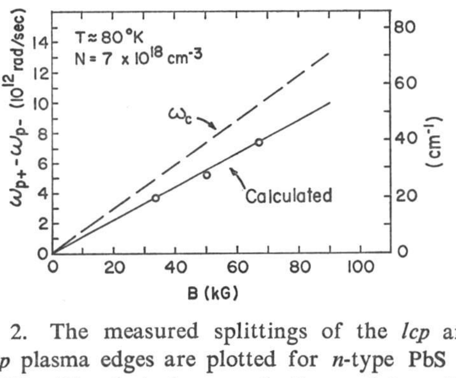

Fig. 2. The measured splittings of the *lcp* and *rcp* plasma edges are plotted for *n*-type PbS at 40°K. The dashed curve is the cyclotron frequency calculated for an effective mass $m_c=0.12$. The splitting calculated, including the interband band-population effects, with the same effective mass is given by the solid curve. If these effects are ignored, then an anomalous value for the effective mass is obtained ($m_c=0.16$).

but not properly identified. Dresselhaus and Mavroides[4] observed such oscillations in the magneto-plasma edge of antimony but refer to theories which only appear to include effects in σ_{xx}. From our calculations it appears that such terms would be small compared to the DHVA terms in $\sigma_{xy}^{(1)}$ and we suggest that the experiment be re-interpreted according to the present model.

§ 4. Conclusions

We have calculated the magneto-conductivity for a simple band model, appropriate to PbS, with a partially populated band. The interband contributions to the *lcp* and *rcp* dielectric constants at frequencies below the band-gap are significantly altered from those for a pure crystal. The resulting changes in the Faraday rotation and the magneto-plasma edges have DHVA-type oscillatory, as well as non-oscillatory components. The experimental observation of the oscillatory component in the Faraday rotation and the non-oscillatory component in the plasma edges are reported.

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DISCUSSION

Smith, S. D.: I wish to comment that the population effect may not be the dominant one in III-V compounds where, the valence bands being degenerate, there is competition between light hole and heavy hole states when the contributions are summed. The experimental effects may then be primarily the effect of blocking on the states near the valence band edge affecting the balance as suggested by Boswarva and Lidiard. The existence of a delicate balance may also account for the non-linear magnetic field dependence which we have recently observed in pure InSb at 5°K.

Mitchell, D. L.: Boswarva and Lidiard did not consider the difference of population in the two sub-bands which are Kramers' degenerate in the absence of a magnetic field. Thus they missed the contributions to $\sigma_{xy}^{(1)}$ which are dominant for the case of the lead salts. These contributions will also be present in the III-V compounds in addition to the usual population blockage terms. The relative size of the two effects remains to be calculated for these compounds. As to Dr. Smith's second comment: I have not seen published data for pure InSb and cannot comment. It is not related to the effects discussed here.

Nishina, Y.: With reference to your work, would you comment on the theoretical work of T. Murao and A. Ebina (J. Phys. Soc. Japan **20** (1965) 997), who derived the expression for the Faraday rotation, taking into account the population effects on the magnetic sublevels?

Mitchell, D. L.: The work of Murao and Ebina has no bearing on the present case since only discrete oscillators were considered. For each oscillator, the $\lim_{\omega\to 0} \sigma_{xy}^{(1)}=0$. This is not true for Bloch electrons in a partially filled band as in the present case. This is discussed by D. L. Mitchell, E. D. Palik and R. F. Wallis: Phys. Rev. Letters **14** (1965) 827.