

## THEORY OF INTERBAND FARADAY ROTATION IN SEMICONDUCTORS

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In the experimental investigation of Faraday rotations in semiconductors, it has been observed that large changes in rotation occur at energies close to the energy gap.<sup>1</sup> In attempting to treat this in InSb, it has been recognized by Stephen and Lidiard<sup>2</sup> that the classical model of bound valence electrons is only qualitative in interpreting this phenomena. Consequently, Lax<sup>3</sup> has proposed a quantum mechanical approach to interpret this in terms of virtual interband transitions associated with the direct transition in this material. It can be shown that similar calculations can be made for the indirect and direct forbidden transitions to calculate the interband Faraday rotation and its dependency on wavelength.

Consider first the indirect case. It is known that the absorption coefficient for simple quadratic bands in the presence of a magnetic field,  $H$ , is given by the following expression<sup>4</sup>:

$$\alpha_i^\pm(\omega, H) = (A/\omega)\hbar^2 \omega_{c1} \omega_{c2} \sum_{n_1 n_2} S(\omega - \omega_{12}^\pm), \quad (1)$$

where  $\omega_{12}^\pm = \omega_g + \omega_p + \omega_{c1}(n_1 + \frac{1}{2}) + \omega_{c2}(n_2 + \frac{1}{2}) \pm \Delta\omega_i$ . Here  $S(\omega - \omega_{12}^\pm)$  is a step function.  $\Delta\omega_i = \gamma_i H$  is the differential frequency shift for left and right circularly polarized waves which propagate along the magnetic field  $H$ . The superscripts  $\pm$ , respec-

tively, correspond to left and right circular waves;  $\omega_g$  is the frequency corresponding to the energy gap,  $\omega_p$  to the phonon energy either in emission (+) or absorption (-).  $\omega_{c1} = eH/m_1c$  is the cyclotron frequency for the valence band and  $\omega_{c2}$  for the conduction band.  $A$  is a constant which involves fundamental parameters, temperature, and matrix elements. We represent  $\gamma_i$  as a phenomenological parameter which can be evaluated for each transition in terms of the selection rules for interband transitions between Landau levels above the energy gap. For simplicity we shall assume that, to the first approximation,  $\gamma_i$  is the same for both left and right circularly polarized waves and is the same for all combinations of quantum numbers,  $n_1$  and  $n_2$ . In the indirect transition where phonon absorption and emission are involved, the selection rules for orbital and magnetic quantum numbers are presumably not conserved.

According to the Kramers-Kronig relationship, the index of refraction at frequency  $\omega$  is

$$n^\pm(\omega) = 1 + \frac{2c}{\pi} \int_0^\infty \frac{\alpha^\pm(\omega') d\omega'}{\omega'^2 - \omega^2}. \quad (2)$$

Hence, the dispersion associated with the absorption coefficient  $\alpha_i^\pm(\omega)$  in terms of the phase constant  $\beta_i^\pm(\omega)$  is

$$\begin{aligned} \beta_i^\pm(\omega) &= \beta_0 + 2A\pi^{-1} \omega \hbar^2 \omega_{c1} \omega_{c2} \int_0^\infty \sum_{n_1 n_2} \frac{S(\omega' - \omega_{12}^\pm) d\omega'}{\omega'(\omega'^2 - \omega^2)} \\ &= \beta_0 + 4A(\pi\omega)^{-1} \hbar^2 \omega_{c1} \omega_{c2} \sum_{n_1 n_2} \ln[\omega_{12}(\omega_{12}^2 - \omega^2)^{-1/2}], \end{aligned} \quad (3)$$

where  $\omega_{12} = \omega_g + \omega_p + \omega_{c1}(n_1 + \frac{1}{2}) + \omega_{c2}(n_2 + \frac{1}{2})$ , and  $\beta_0 = \omega/c$ .

The above expression applies in  $\omega$  both below and above the energy gap. For the latter case,  $\beta_i^\pm(\omega)$  should indicate the structure associated

with the transition between each pair of magnetic levels  $n_1$  and  $n_2$ . For a weak magnetic field, and at frequencies well below the energy gap,  $\Delta\omega_i \ll \omega_{12}^\pm - \omega$ . Then  $\beta_i^\pm(\omega)$  may be expanded

with respect to  $\Delta\omega_i$  and the Faraday rotation angle per unit length,  $\theta_i$ , given by  $\theta_i = (\beta_i^+ - \beta_i^-)/2$  gives the following result:

$$\theta_i = -\frac{2}{\pi} A \hbar^2 \omega_{c1} \omega_{c2} \omega \sum_{n_1 n_2} \frac{\Delta\omega_i}{(\omega_{12}^2 - \omega^2) \omega_{12}} \quad (4)$$

For the frequency  $\omega$  well below the gap, the summation merely represents the tail of the dispersion for each of the transitions and, hence, to the first approximation should not differ from the limit at zero magnetic field. Thus, the summation can be evaluated to this limit by integrating to obtain the expression for the Faraday rotation  $\theta_i$  as follows:

$$\theta_i = -\frac{2}{\pi} A \hbar^2 \frac{\Delta\omega_i}{\omega} \left[ \omega \ln \frac{g^2 - \omega^2}{\omega^2} + \omega \ln \frac{g + \omega}{g - \omega} \right], \quad (5)$$

where  $\lim_{H \rightarrow 0} \omega_{12} = \omega_g$ . The above expression approaches  $-(2/\pi) A \hbar^2 \Delta\omega_i \ln 2$  as  $\omega \rightarrow \omega_g$ , and asymptotically reduces to zero as  $\omega$  becomes small compared to  $\omega_g$ .

In a similar way we can evaluate the Faraday rotation due to the direct transition from the well-known absorption coefficient  $\alpha_d^\pm(\omega, H)$ ,<sup>4</sup> where

$$\alpha_d^\pm = \frac{1}{2} B (\hbar\omega/c) \sum_n (\omega - \omega_n^\pm)^{-1/2}. \quad (6)$$

Here  $\omega_n^\pm = \omega_g + (n + \frac{1}{2})\omega_c \pm \Delta\omega_d$ ;  $B = 2e^2(2\mu)^{3/2} \times (\eta m^2 c)^{-1} \hbar^{-1/2} |\vec{P}_{12} \cdot \vec{e}|^2$ ,  $\mu = m_1 m_2 / (m_1 + m_2)$  = reduced mass,  $\omega_c = \omega_{c1} + \omega_{c2}$ ,  $m$  = free electron mass,  $\eta$  = index of refraction, and  $\vec{P}_{12}$  = momentum matrix. The final result for frequency  $\omega < \omega_g$  was found to be

$$\theta_d = \frac{B \hbar \omega}{4\omega c} \sum_n \Delta\omega_d [2\omega_n^{-3/2} - (\omega_n + \omega)^{-3/2} - (\omega_n - \omega)^{-3/2}], \quad (7)$$

where  $\omega_n = \omega_g + (n + \frac{1}{2})\omega_c$ . In this case  $\Delta\omega_d = \gamma_d H$  can be in principle evaluated for each transition, since the selection rules for the normal magnetic level structures have been worked out.<sup>5</sup> The sign of the value of  $\gamma_d$  for such direct transitions will then be determined by the admixture of orbital states as well as effective  $g$  values in the presence of spin-orbit coupling. We shall again assume that  $\Delta\omega_d$  is the same for each transition. The summation was approximated by an integral over  $n$  in the limit of weak magnetic field, with the result

$$\theta_d = \frac{1}{2} B \hbar (\Delta\omega_d / \omega) [2\omega_g^{-1/2} - (\omega_g + \omega)^{-1/2} - (\omega_g - \omega)^{-1/2}]. \quad (8)$$

The above results in Eqs. (5) and (8) can read-

ily give the semiquantitative explanation of the interband Faraday rotation observed by Hartmann and Kleman<sup>1</sup> in Ge where the direct and indirect energy gaps are close to one another. Assuming that  $\Delta\omega_i$  is negative for a positive value of magnetic field, our result indicates that the Faraday rotation for the indirect transition has the same sign, i.e., positive, as that for free carrier. This sign is consistent with the experimental result.<sup>1,3</sup>  $\Delta\gamma_d$  must be positive, since the interband Faraday rotation due to the direct transition is negative. Its magnitude becomes very large as the incident photon energy approaches the gap energy. Both from the theoretical and experimental curves in Figs. 1 and 2, which are

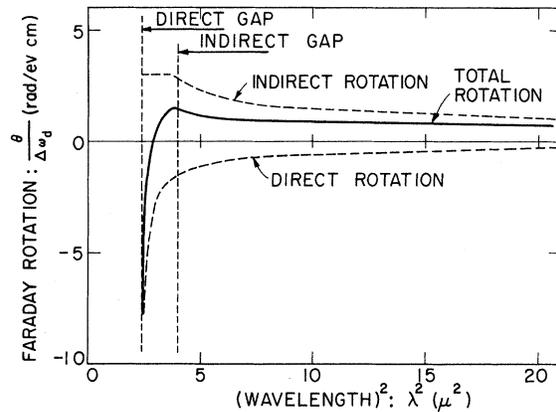


FIG. 1. Theoretical curve of Faraday rotation in intrinsic Ge at room temperature. The dashed lines are individual contributions of indirect and direct interband virtual transitions. The solid line is the combined total rotation. The magnitude of the dispersion was evaluated from the absorption coefficients for both transitions from the experimental data by W. C. Dash and R. Newman [Phys. Rev. **99**, 1151 (1955)] at zero magnetic field.

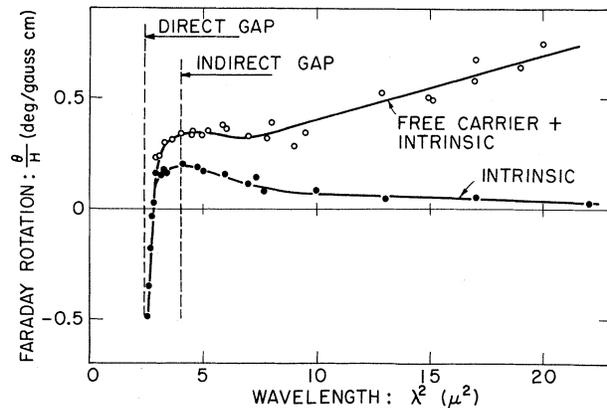


FIG. 2. Experimental curve of Faraday rotation in intrinsic Ge at room temperature by Hartmann and Kleman.<sup>1</sup>

similar in shape, it is apparent that a strong singularity in Faraday rotation must correspond to the direct transition consistent with the theoretical result of Eq. (8).

The forbidden direct transition can be treated in an analogous manner. The two bands have the same symmetry, so that no transition is allowed at  $k=0$ . In this case the absorption coefficient is given by the expression,

$$\alpha_f^\pm = \frac{3}{8} \left( \frac{\hbar\omega_c}{2} \right)^2 \frac{C}{\omega} \sum_n (n+1) [(\omega - \omega_1^\pm)^{-1/2} + (\omega - \omega_2^\pm)^{-1/2}], \quad (9)$$

where  $\omega_1^\pm = \omega_g + (n + \frac{1}{2})\omega_c + \omega_{c1} \pm \Delta\omega_f$ , (and similarly for  $\omega_2^\pm$ ), and  $C = \frac{2}{3}e^2(\eta m^2 c)^{-1}\hbar^{-7/2} |\bar{M}_{12}|^2 (2\mu)^{5/2}$ ,  $\bar{M}_{12}$  being the momentum matrix. Following the steps indicated previously, we obtain the result for the Faraday rotation angle per unit length:

$$\theta_f = \frac{3}{16} (\hbar\omega_c)^2 (C/\omega) \sum_n (\Delta\omega_f) (n+1) [\omega_1^{-3/2} - \frac{1}{2}(\omega_1 + \omega)^{-3/2} - \frac{1}{2}(\omega_1 - \omega)^{-3/2} + \omega_2^{-3/2} - \frac{1}{2}(\omega_2 + \omega)^{-3/2} - \frac{1}{2}(\omega_2 - \omega)^{-3/2}]. \quad (10)$$

In the limit of weak magnetic fields we obtain, by integrating Eq. (10),

$$\theta_f \approx \frac{3}{4} \hbar^2 C (\Delta\omega_f/\omega) [-2\omega_g^{1/2} + (\omega_g + \omega)^{1/2} + (\omega_g - \omega)^{1/2}]. \quad (11)$$

This result has some interesting application to *p*-type Ge, where such transitions have been observed between the split valence bands. The two prominent absorptions observed at room temperature at approximately 3 and 5 microns should give rise to peaks in the curve of Faraday rotation as shown in Fig. 3. These correspond to the transitions between  $P_{1/2}$  and the degenerate  $P_{3/2}$

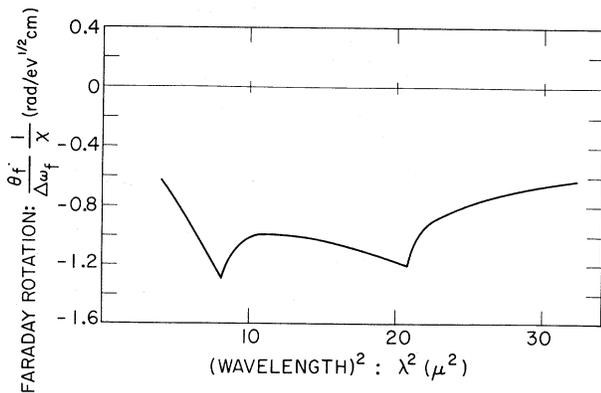


FIG. 3. Theoretical curve of Faraday rotation due to forbidden direct transitions in Ge at room temperature. The relative magnitudes of absorption coefficients and position of peaks were determined from the experimental curve of Kaiser, Collins, and Fan.<sup>6</sup>  $\chi = \frac{3}{4} \hbar^2 C$  in Eq. (11).

bands. The sign of rotation is not known from theory. We have assumed  $\Delta\omega_f$  to be positive, giving negative values of  $\theta$  as shown in Fig. 3. However, experimental results will determine the sign. It is necessary to use doped samples to observe these transitions. Hence, the contribution from the free-carrier absorption would be superimposed. At low temperatures where many of the light-hole states are filled, only the transition to the heavy-hole states about 4 microns would be observable.<sup>6</sup> At such temperatures and in very high magnetic fields, it is possible that only a few transitions can be seen, so that the width of the peak in the Faraday rotation would be small, giving rise to fine structures. In this case the Faraday rotation should be expressed in terms of Eq. (10) rather than Eq. (11).

The theory is only approximate, since it makes a number of assumptions. One assumption is that the bands are quadratic and extend to large  $k$  values without changing the curvature. This is incorrect since the dispersion or phase constant obtained by Kramers-Kronig relationship at zero magnetic field would diverge for the indirect and the forbidden direct transitions. This contradiction can be avoided by using the nonparabolic representation of the bands, when they are known, as in InSb. The wavelength dependence of the Faraday rotation would be modified for nonparabolic bands. The theory has also neglected the losses which can be taken into account phenomenologically in the final expressions. This

has been done for the direct interband transitions above the energy gap. For more accurate analysis of the Faraday rotation in *p*-type Ge, it is also necessary to take statistics into account since the dispersion must be weighted by the number of empty states. In spite of these assumptions, however, the above approximate results seem to be reasonable because they can explain the major features of the experimental observations, whereas the classical theory for bound electrons cannot. Lastly, in order to account for the proper sign of the interband Faraday rotation, it is necessary to consider in detail the individual magnetic levels of the two sets of the bands involved in transition, including spin, particularly in the presence or absence of spin-orbit splitting. The analysis would presumably account for the apparent difference between the observation in GaAs as contrasted to InSb and InAs.<sup>1</sup>

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### HYPERFINE INTERACTIONS IN THE GROUND STATE AND FIRST EXCITED STATE OF Dy<sup>161</sup> IN DYSPROSIUM IRON GARNET\*

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The hyperfine Zeeman splittings in the recoil-free absorption spectrum<sup>1</sup> of the 26-keV  $\gamma$  rays from Dy<sup>161m</sup> ( $\tau_{1/2} \sim 3 \times 10^{-8}$  sec)<sup>2</sup> in Dy<sup>161</sup> situated in dysprosium iron garnet (DIG) have been observed. This is possible since in the rare earth iron garnets (5Fe<sub>2</sub>O<sub>3</sub>·3M<sub>2</sub>O<sub>3</sub>) the sublattice of rare earth ions below the Curie temperature shows a temperature-dependent spontaneous magnetization.<sup>3</sup> Nearly all the rare earth elements can be incorporated in iron garnets and since an appreciable recoil-free efficiency is expected at liquid air temperature for the  $\gamma$  transition from the first excited state of many rare earth nuclei, the method is of general application. Values for the effective magnetic field at the nucleus ( $H_{\text{eff}}$ ) and the magnetic and quadrupole moments of the ground state and

first excited state of Dy<sup>161</sup> are derived. At 85°K a large nuclear quadrupole interaction has been observed, but at 300°K the quadrupole interaction is much smaller. This strongly temperature-dependent interaction is interpreted as being largely due to the average electric field gradient produced by the partially aligned orbitals of the 4f electron shell and correlated with the spontaneous magnetization (and  $H_{\text{eff}}$ ) within each domain. This gradient will have an axis of symmetry parallel to  $H_{\text{eff}}$ .

The recoil-free absorption spectrum of polycrystalline DIG, isotopically enriched in Dy<sup>161</sup>, was measured using a source of Dy<sup>161m</sup> which showed an unsplit emission line of relatively narrow width. This was prepared as follows: 6.7-day Tb<sup>161</sup> was separated by ion exchange