PHYSICAL REVIEW VOLUME 134, NUMBER 1A 6 APRIL 1964

Quantum Theory of Interband Faraday and Voigt Effects

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(Received 17 June 1963; revised manuscript received 14 October 1963)

A quantum-mechanical analysis of the Faraday rotation and the Voigt effect has been carried out for both the oscillatory and long-wavelength regions. Expressions have been developed for these effects from the off-diagonal and diagonal components, respectively, of the conductivity tensor; the latter has been obtained in the form of the Kramers-Heisenberg dispersion relations through the use of first-order time-dependent perturbation theory. The results, which have been calculated for a simplified two-band model, are generalized to apply in the high-field case as well as the low-field limit. Through the introduction of a phenomenological relaxation time, τ, line shapes have been calculated for both the direct and indirect transition for the Faraday and Voigt effects. These have been obtained as a function of frequency for various values of magnetic field and relaxation times. The results obtained enable the evaluation of g factors from experimantal line shapes.

INTRODUCTION

RECENTLY, a number of experimental and theoretical papers have been reported on the Faraday rotation due to interband transitions, particularly in the low-frequency region.[1,2] These results have indicated that the earlier theoretical developments were either incomplete or inappropriate as far as the quantitative interpretation of these results is concerned. The first approach to the problem was a qualitative one by Stephen and Lidiard[3] who made the prediction that some sort of singularity in the Faraday rotation would be observed near the energy gap. They based their prediction on an expression obtained for a single classical oscillator of the form

$$\theta \sim \frac{\omega^2 \omega_c}{(\omega_0{}^2 - \omega^2)^2}, \tag{1}$$

where ω_c is the cyclotron frequency, ω_0 is the bound oscillator frequency, ω the frequency of the infrared or optical wave, and θ the Faraday angle. Although qualitatively Eq. (1) properly embodies the physical phenomenon involved, it was shortly pointed out by Lax[4] that this expression was inappropriate and that in order to properly explain the singularity, which was subsequently obtained experimentally by Brown[5] and also by Walton and Moss,[6] it was necessary to take into account the band properties of the semiconductor and to obtain the dispersion corresponding to the absorption near the energy gap. The expression obtained on this basis was shown, for the dominant term, to be of the form[4]

$$\theta_d \sim \frac{\omega_c \gamma H}{\omega} \sum_n (\omega_n - \omega)^{-3/2}, \quad \omega > \omega_g, \tag{2a}$$

$$\theta_d \sim \frac{\gamma H}{\omega} (\omega_g - \omega)^{-1/2}, \quad \omega < \omega_g. \tag{2b}$$

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† Operated with support from the U. S. Army, Navy, and Air Force.

‡ Supported by the U. S. Air Force Office of Scientific Research.

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2 Theoretical: M. J. Stephen and A. D. Lidiard, Phys. Chem. Solids **9**, 43 (1959); H. S. Bennett and E. A. Stern, University of Maryland Technical Report No. 197, 1960 (unpublished); B. Lax and Y. Nishina, Phys. Rev. Letters **6**, 464 (1961); M. Suffczynski, Proc. Phys. Soc. (London) **77**, 1042 (1961); I. M. Boswarva, R. E. Howard, and A. D. Lidiard, Proc. Roy. Soc. (London) **A269**, 125 (1962); J. Kolodziejczak, B. Lax, and Y. Nishina, Phys. Rev. **128**, 2655 (1962).

3 M. J. Stephen and A. D. Lidiard, Phys. Chem. Solids **9**, 43 (1959).

4 B. Lax, *Proceedings of the International Conference on Semiconductor Physics, 1960* (Czechoslovakian Academy of Sciences, Prague, 1961), p. 321.

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6 A. K. Walton and T. S. Moss, Proc. Phys. Soc. (London) **78**, 1393 (1961).

These expressions are appropriate for photon energies near the energy gap. Equation (2a) represents the oscillatory case just above the gap and in essence is the correct result if the less dominant terms are neglected. However, for the region below the gap it was soon recognized that the single term (2b) was inadequate to describe the total dispersion and that additional terms were required, particularly in the low-frequency limit. Lax and Nishina[7] obtained such expressions through the use of the Kramers-Kronig relations. However, it was subsequently recognized on theoretical grounds by Kolodziejczak, Lax, and Nishina[8] (KLN) and also pointed out by Boswarva, Howard, and Lidiard[9] (BHL) that these results were in error; the expressions derived were essentially for the diagonal components of the dispersion tensor and therefore gave an (incorrect) low-frequency behavior that went as λ^{-1}. Independently, subsequent experimental observations of Smith, Pidgeon, and Prosser,[10] Piller and Potter,[11] and also of Moss, Walton, and Ellis[12] clearly demonstrated a λ^{-2} behavior. A (phenomenological) semiclassical theory was subsequently developed by KLN[8] which did exhibit the λ^{-2} low-frequency dependence which satisfied the appropriate Kramers-Kronig relations and exhibited the correct symmetry properties. At the same time, Boswarva, Howard, and Lidiard[9] developed the quantum-mechanical Kramers-Heisenberg approach to the problem. Unfortunately, they made an assumption with respect to the momentum matrix elements which led to results which did not satisfy the low-frequency limit and consequently they they added a correction term.

The object of the present paper is to modify and extend the BHL treatment and to obtain expressions for the Faraday rotation which are of the same form in the long-wavelength limit as the KLN semiclassical expressions. To do this we shall follow the BHL approach and notation to the point of departure which is essentially that the matrix elements for the left and right circularly polarized waves, in the presence of a magnetic field, differ by the same order of magnitude as the eigenvalues and are not equal as assumed by BHL. However, it should be pointed out that an assumption still must be made to obtain quantitative results, and this will subsequently be discussed. We shall also use the quantum treatment to obtain expressions for the Voigt effect and to obtain the line shapes in the oscillatory region for both the Faraday rotation and the Voigt phase shift.

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[8] J. Kolodziejczak, B. Lax, and Y. Nishina, Phys. Rev. **128**, 2655 (1962).

[9] I. M. Boswarva, R. E. Howard, and A. D. Lidiard, Proc. Roy. Soc. (London) **A269**, 125 (1962).

[10] S. D. Smith, C. R. Pidgeon, and V. Prosser, *Proceedings of the International Conference on the Physics of Semiconductors, Exeter, 1962* (The Institute of Physics and The Physical Society, London, 1962), p. 301.

[11] H. Piller and R. F. Potter, Phys. Rev. Letters **9**, 203 (1962).

[12] T. S. Moss, A. K. Walton, and B. Ellis, *Proceedings of the International Conference on the Physics of Semiconductors, Exeter* (The Institute of Physics and The Physical Society, London, 1962), p. 295.

FARADAY EFFECT

From the time-dependent perturbation theory, the current density vector may be obtained and it can then be shown that the conductivity tensor for all transitions is given by

$$\sigma_{ii}=-\frac{ie^2}{m^2\hbar}\sum_k\sum_{k'}\frac{1}{\omega_{kk'}}\left\{\frac{P_{kk'}{}^iP_{k'k}{}^i}{\omega+\omega_{kk'}}+\frac{P_{kk'}{}^iP_{k'k}{}^i}{\omega-\omega_{kk'}}\right\}, \quad (3a)$$

$$\sigma_{ij}=\frac{ie^2}{m^2\hbar\omega}\sum_k\sum_{k'}\left\{\frac{P_{kk'}{}^iP_{k'k}{}^j}{\omega+\omega_{kk'}}-\frac{P_{kk'}{}^jP_{k'k}{}^i}{\omega-\omega_{kk'}}\right\}, \quad (3b)$$

for the diagonal and off-diagonal components, respectively, where $P_{kk'}$ is the momentum matrix for the transitions between states k and k' and includes both the magnetic-field and the spin-orbit coupling

$$\mathbf{P}=\mathbf{p}+e\mathbf{A}+(1/2m^2)(\mathbf{S}\times\nabla V). \quad (4)$$

In order to show the appropriate form of the off-diagonal component of the conductivity tensor in the presence of the magnetic field, we use the usual approximation that the conduction current is small compared with the displacement current ($\sigma\ll\omega\epsilon$) and hence the Faraday rotation is given to a good approximation by the expression

$$\theta\approx -K_e^{1/2}\sigma_{xy}/2c\epsilon, \quad (5)$$

where σ_{xy} is the dispersive or nondissipative part of the conductivity. Here K_e is the dielectric constant and ϵ is the permittivity of the medium; mks units are used throughout. We assume that the medium is gyrotropic, for which we have $\sigma_{xy}=-\sigma_{yx}$. Using (3b) and the above symmetry property, we can interchange $P_{kk'}{}^xP_{k'k}{}^y$ and $-P_{kk'}{}^yP_{k'k}{}^x$ to obtain for θ

$$\theta=\frac{iK_e{}^{1/2}e^2}{m^2\hbar\epsilon c}\sum_k\sum_{k'}\frac{P_{kk'}{}^xP_{k'k}{}^y}{\omega_{kk'}{}^2-\omega^2}. \quad (6)$$

The validity of the step leading to Eq. (6) hinges upon the choice that the model exhibits cubic symmetry, since when one performs a 90° rotation about the z axis, which takes $x\to y$ and $y\to -x$, one also transforms the k and k'. However, because of the cubic symmetry, the energy denominators remain unchanged. As shown by BHL, the above expression can be transformed by means of the introduction of momentum operators corresponding to polarization vectors of two circularly polarized components rotating in a clockwise and counterclockwise sense, respectively (i.e., $P^\pm=P^x\pm iP^y$); substitution for P^x and P^y and the application of Hermiticity conditions gives

$$|P_{kk'}{}^+|^2-|P_{kk'}{}^-|^2=2i(P_{kk'}{}^yP_{k'k}{}^x-P_{kk'}{}^xP_{k'k}{}^y). \quad (7)$$

Substitution of Eq. (7) into (6) subject to the interchange of $-P_{kk'}{}^{y}P_{k'k}{}^{x}$ and $P_{kk'}{}^{x}P_{k'k}{}^{y}$ as just discussed gives the BHL result; however, we shall depart from the representation they obtained as follows: We write[13]

$$\theta=\frac{-K_e^{1/2}e^2}{4m^2\hbar\epsilon c}\sum_k\sum_{k'}\left\{\frac{|P_{kk'}{}^{+}|^2}{(\omega_{kk'}{}^{+})^2-\omega^2}-\frac{|P_{kk'}{}^{-}|^2}{(\omega_{kk'}{}^{-})^2-\omega^2}\right\},\quad(8)$$

where we have a different denominator for each of the two sets of transitions since the eigenvalues associated with the two operators differ in accordance with the selection rules determined by the matrices. The matrix element of $P_{kk'}{}^{+}$ is zero for one member, and that of $P_{kk'}{}^{-}$ is zero for the other member of the "$\pm$" pair of excitations. The $\omega_{kk'}{}^{\pm}$ are defined by

$$\omega_{kk'}{}^{\pm}=\omega_{kk'}\pm\gamma H\,,\quad(9)$$

where, for the direct transition

$$\omega_{kk'}\rightarrow\omega_{kk}=\omega_g+(n+\tfrac{1}{2})\omega_c+p_z{}^2/2\mu\hbar\,,\quad(9\text{a})$$

and for the indirect transition

$$\omega_{kk'}=\omega_g+(n_1+\tfrac{1}{2})\omega_{c_1}+(n_2+\tfrac{1}{2})\omega_{c_2}\pm k\theta/\hbar+p_{1_z}{}^2/2m_1\hbar+p_{2_z}{}^2/2m_2\hbar\,,\quad(9\text{b})$$

where ω_g is the frequency corresponding to the energy gap, the ω_c are the cyclotron frequencies, $\gamma=(g_c+g_v)\times(\mu_B/2\hbar)$ is the appropriate effective gyromagnetic ratio for the transition between valence and conduction bands, μ_B is the Bohr magneton, and $k\theta/\hbar$ represents the phonon involved in the indirect transition.

In accordance with the above representation for the two sets of eigenvalues for left and right circularly polarized waves we can show that the matrix elements P^{+} and P^{-} *also differ by the same ratio*. It has been shown theoretically by BHL and it is also known from experiment that the correct theory must give $\theta\rightarrow 0$ as $\omega\rightarrow 0$. Hence, if we set $\omega=0$ in Eq. (5), we obtain

$$\sum_k\sum_{k'}\frac{|P_{kk'}{}^{+}|^2}{(\omega_{kk'}{}^{+})^2}=\sum_k\sum_{k'}\frac{|P_{kk'}{}^{-}|^2}{(\omega_{kk'}{}^{-})^2}\,.\quad(10)$$

We shall further argue for our model[14] that the above equality holds for *each* set of transitions kk' so that

$$\frac{|P_{kk'}{}^{+}|}{\omega_{kk'}{}^{+}}=\frac{|P_{kk'}{}^{-}|}{\omega_{kk'}{}^{-}}\equiv\frac{|P_{kk'}|}{\omega_{kk'}}\,.\quad(11)$$

The above is quite reasonable from the following physical arguments: We can consider each set of levels as discrete, since the energy bands are quasicontinuous, and that the transition between two complementary levels is equivalent to the classical situation treated by KLN. Therefore, from the correspondence principle, the Faraday rotation for *each* of these transitions must also go to zero, which then justifies the equality (11).

As the magnetic field is increased the matrix elements also increase linearly in proportion to the field. The above expression essentially compares the matrix elements about the mean of the two circularly polarized components; it is not an expansion as a function of the total magnetic field but only an increment about each magnetic level which is split by the field. In this regard it differs from the treatment of Bennett and Stern[15] and of Roth.[16]

Substituting (11) into (8), and after some algebraic manipulation, we obtain

$$\theta=\frac{-K_e^{1/2}e^2}{4m^2\hbar\epsilon c}\sum_k\sum_{k'}|P_{kk'}|^2\times\frac{\omega^2}{\omega_{kk'}{}^2}\left(\frac{1}{(\omega_{kk'}{}^{+})^2-\omega^2}-\frac{1}{(\omega_{kk'}{}^{-})^2-\omega^2}\right).\quad(12)$$

We can examine Eq. (12) in the limit of small γH; it reduces to

$$\theta\rightarrow\frac{K_e^{1/2}e^2}{m^2c\hbar\epsilon}\sum_k\sum_{k'}\frac{|P_{kk'}|^2}{\omega_{kk'}}\frac{\omega^2(\gamma H)}{(\omega_{kk'}{}^2-\omega^2)^2}\,.\quad(13)$$

Here $|P_{kk'}|^2/\omega_{kk'}$ is, aside from a constant factor, just the oscillator strength. Equation (13) is of the identical form as that obtained from the semiclassical theory of KLN and disagrees with the results of BHL based on the assumption that $|P_{kk'}{}^{+}|\approx|P_{kk'}{}^{-}|$. Furthermore, BHL, in order to obtain the proper low-frequency limit, i.e., $\theta\sim\omega^2$, had to add a correction term to satisfy this criterion. The above result (13), when evaluated for a simple parabolic conduction and valence band by integrating over the quasicontinuous values of the momentum, gives five terms; these are given for the direct and indirect transitions in the paper by KLN.

We now return to the quantum-mechanical expression of Eq. (12). Expansion in partial fractions gives

$$\theta=\frac{K_e^{1/2}e^2\omega}{8m^2\hbar c\epsilon}\sum_k\sum_{k'}|P_{kk'}|^2\left\{\frac{8\gamma H\omega}{[\omega^2-(\gamma H)^2]^2\omega_{kk'}}-\frac{1}{(\omega-\gamma H)^2(\omega_{kk'}-\omega+\gamma H)}+\frac{1}{(\omega+\gamma H)^2(\omega_{kk}{}'+\omega+\gamma H)}+\frac{1}{(\omega+\gamma H)^2(\omega_{kk'}-\omega-\gamma H)}-\frac{1}{(\omega-\gamma H)^2(\omega_{kk'}+\omega-\gamma H)}\right\}.\quad(14)$$

[13] This expression was independently derived by H. S. Bennett (private communication).

[14] As regards this assumption, L. M. Roth has made the following comments in a private communication. "The ω^2 dependence comes from the cancellation between the energy shifts γH and the H dependence of the matrix elements. The present paper uses a model for which the cancellation comes about for each transition. This is analogous to the work of Rosenfeld [see Van Vleck, *Electric and Magnetic Susceptibilities* (Oxford University Press, New York, 1932), p. 368]. BHL assume that the cancellation comes about entirely from other bands, which seems less reasonable. At any rate, the correct answer would probably lie in between."

[15] H. S. Bennett and E. A. Stern, University of Maryland Technical Report No. 197, 1960 (unpublished).

[16] L. M. Roth, Phys. Rev. **133**, A542 (1964).

Direct Transition

For the direct transition, we have $\omega_{kk'}\rightarrow\omega_{kk}=\omega_g+(n+\frac{1}{2})\omega_c+(p_z^2/2\mu\hbar)$. To obtain the contribution to the rotation for a transition between a given pair of Landau levels we must integrate over p_z. We have

$$\sum_k\rightarrow\frac{1}{h^3}\int d^3p\rightarrow\sum_n\frac{\mu\omega_c}{2\pi^2\hbar^2}\int dp_z,$$

where the factor $\omega_c=eH/\mu c$ essentially accounts for the density of states. Performing the integration over $\int dp_z$ we obtain for a single Landau transition

$$\theta_{d\,(\text{Landau})}=\frac{K_e^{1/2}e^2\omega\omega_c(2\mu)^{3/2}}{128\pi m^2c\epsilon\hbar^{5/2}}|P_{kk}|^2\left\{\frac{8\gamma H\omega}{[\omega^2-(\gamma H)^2]^2\omega_n^{1/2}}-\frac{1}{(\omega-\gamma H)^2(\omega_n-\omega+\gamma H)^{1/2}}\right.$$
$$\left.+\frac{1}{(\omega+\gamma H)^2(\omega_n+\omega+\gamma H)^{1/2}}+\frac{1}{(\omega+\gamma H)^2(\omega_n-\omega-\gamma H)^{1/2}}-\frac{1}{(\omega-\gamma H)^2(\omega_n+\omega-\gamma H)^{1/2}}\right\},\quad(15)$$

where

$$\omega_n=\omega_g+(n+\tfrac{1}{2})\omega_c.$$

To obtain the line shape in the vicinity of a Landau level we assume $\omega\gg\gamma H$, extract the terms with the singularities since these will be the terms making the major contributions near the given Landau level, and then introduce a phenomenological relaxation time τ, by letting $\omega\rightarrow\omega-i/\tau$; the justification for this last step can be obtained from the introduction of a damping term in the classical equation of motion for the bound oscillator.

$$\theta_{d\,(\text{Landau})}=\frac{K_e^{1/2}e^2\tau^{1/2}(2\mu)^{3/2}\omega_c}{128\pi m^2c\epsilon\hbar^{5/2}\omega}|P_{kk}|^2\,\text{Re}\left\{\frac{1}{[(\omega_n-\omega-\gamma H)\tau+i]^{1/2}}-\frac{1}{[(\omega_n-\omega+\gamma H)\tau+i]^{1/2}}\right\},\quad(16)$$

letting $X=(\omega_n-\omega)\tau$ and $Y=\gamma H\tau$, we finally obtain

$$\theta_{d\,(\text{Landau})}=A\left\{\frac{1}{[(X-Y)^2+1]^{1/2}}\{[(X-Y)^2+1]^{1/2}+(X-Y)\}^{1/2}\right.$$
$$\left.-\frac{1}{[(X+Y)^2+1]^{1/2}}\{[(X+Y)^2+1]^{1/2}+(X+Y)\}^{1/2}\right\},\quad(17)$$

where

$$A=\frac{K_e^{1/2}e^2\omega_c\mu^{3/2}\tau^{1/2}}{64\pi m^2c\epsilon\hbar^{5/2}\omega}|P_{kk}|^2.\quad(17\text{a})$$

In Figs. 1(a) and (b), we have plotted the expression in the braces to obtain contributions to the line shapes in the vicinity of the frequency corresponding to the transition between the given Landau level and its complement, for various values of $\gamma H\tau$.

If one wishes to be able to compare experimental data with the theoretically calculated line shapes, then it is necessary to evaluate the background rotation since it will in general contain terms which will contribute to the rotation, and hence to the line shapes in the vicinity of the Landau level. The evaluation of the background rotation involves summing over all the Landau levels for the expression in Eq. (15). Now $\sum_n\rightarrow\int dn\rightarrow 1/\omega_c\int dx$, where we define $x=(n+\frac{1}{2})\omega_c$. Performing the integration then gives us the result

$$\theta_{d\,(\text{background})}=\frac{K_e^{1/2}e^2(2\mu)^{3/2}\omega}{64\pi m^2c\epsilon\hbar^{5/2}}|P_{kk}|^2\left\{\frac{-8\gamma H\omega}{[\omega^2-(\gamma H)^2]^2}\omega_g^{1/2}+\frac{1}{(\omega-\gamma H)^2}(\omega_g-\omega+\gamma H)^{1/2}\right.$$
$$\left.-\frac{1}{(\omega+\gamma H)^2}(\omega_g+\omega+\gamma H)^{1/2}-\frac{1}{(\omega+\gamma H)^2}(\omega_g-\omega-\gamma H)^{1/2}+\frac{1}{(\omega-\gamma H)^2}(\omega_g+\omega-\gamma H)^{1/2}\right\}.\quad(18)$$

In the vicinity of the energy gap we assume $\omega\gg\gamma H$, extract the dominant terms, and let $\omega\rightarrow\omega-i/\tau$. This leaves us with

$$\theta_{d\,(\text{background})}=\frac{K_e^{1/2}e^2(2\mu)^{3/2}}{64\pi m^2 c\epsilon\hbar^{5/2}\omega\tau^{1/2}}|P_{kk}|^2\,\text{Re}\{[(\omega_g-\omega-\gamma H)\tau+i]^{1/2}-[(\omega_g-\omega+\gamma H)\tau+i]^{1/2}\} \tag{19}$$

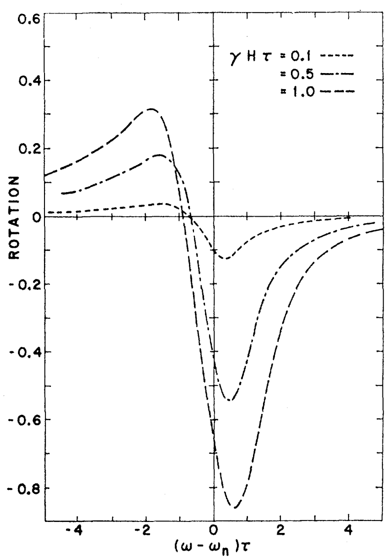
and letting $X=(\omega_g-\omega)\tau$ and $Y=\gamma H\tau$, we obtain

$$\theta_{d\,(\text{background})}=\frac{2A}{\omega_c\tau}\{\{[(X+Y)^2+1]^{1/2}+(X+Y)\}^{1/2}-\{[(X-Y)^2+1]^{1/2}+(X-Y)\}^{1/2}\}. \tag{20}$$

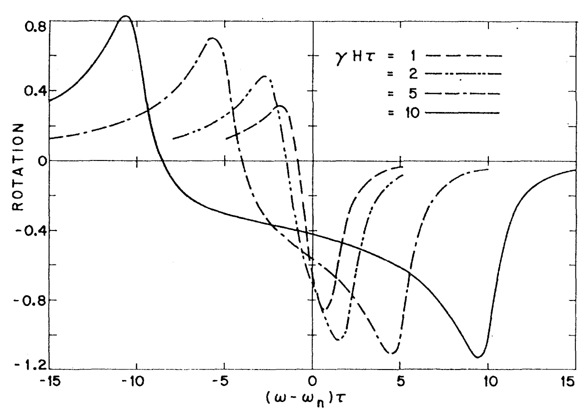
We have plotted the expression in the braces in (20) in Figs. 2(a) and 2(b) with $Y=\gamma H\tau$ as parameter. We note from the $(\omega_c\tau)^{-1}$ in (20) that as τ becomes large, the *relative* contribution of the background to that due to the single Landau level, becomes smaller. Furthermore, in most experimental arrangements at high fields where line-shape study is meaningful, we would expect $\omega_c\tau$ to be of ~ 100; consequently, as a comparison of Figs. 1(a) and 1(b) with 2(a) and 2(b) shows, the background may be neglected compared with the contribution from the single Landau level transition. Hence in the vicinity of the energy gap the first singularity is due to the first Landau transition.

Direct Transition—Long-Wavelength Limit

To obtain the behavior of the Faraday rotation due to the direct transition in the long-wavelength limit we must sum over the contributions from transitions to all Landau levels and hence must start with Eq. (18). In this limit,

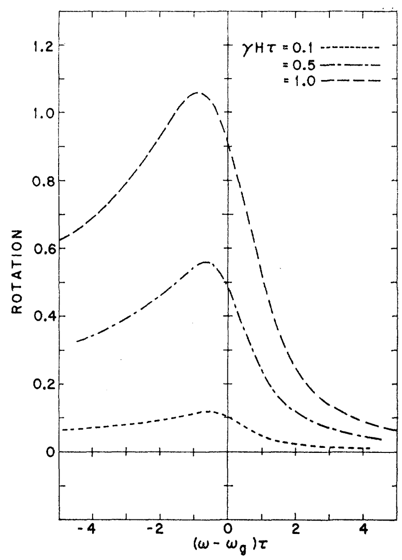


(a)

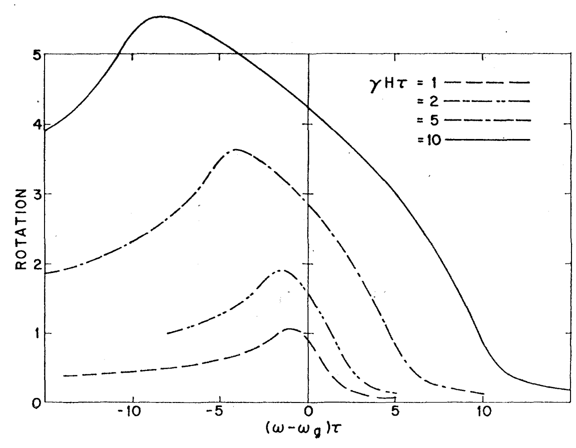


(b)

FIG. 1. (a), (b) Plot of the line shapes for the Faraday rotation direct transition between a given pair of Landau levels as a function of frequency for different values of $\gamma H\tau$. The constant A of Eq. (17a) has been normalized to unity.



(a)



(b)

FIG. 2. (a), (b) Contribution to the Faraday rotation direct transition line shape due to background. The ordinates of the figures must be multiplied by $2/\omega_c\tau$ to permit comparison with Figs. 1 (a) and (b).

however, we have $\omega_g \gg \omega$, γH. Letting $x=(1/\omega_g)(\omega+\gamma H)$; $y=(1/\omega_g)(\omega-\gamma H)$, we obtain

$$\theta_d \underset{\omega\to 0}{\longrightarrow} \frac{K_e^{1/2}e^2(2\mu)^{3/2}\omega\omega_g^{1/2}}{64\pi m^2 c\epsilon\hbar^{5/2}}|P_{kk}|^2\left\{\frac{-8\gamma H\omega}{[\omega^2-(\gamma H)^2]^2}+\frac{1}{(\omega_g y)^2}[(1-y)^{1/2}+(1+y)^{1/2}]-\frac{1}{(\omega_g x)^2}[(1+x)^{1/2}+(1-x)^{1/2}]\right\}. \tag{21}$$

Now x, $y\ll 1$; hence we can expand (21). When we do this we obtain

$$\theta_d \underset{\omega\to 0}{\longrightarrow} \frac{5}{1024\pi}\left(\frac{K_e^{1/2}e^2(2\mu)^{3/2}}{m^2 c\epsilon\hbar^{5/2}}\right)|P_{kk}|^2\frac{\omega^2\gamma H}{\omega_g^{7/2}}. \tag{22}$$

Indirect Transition

For the indirect transition we have

$$\omega_{kk'}=\omega_g+(n_1+\tfrac{1}{2})\omega_{c_1}+(n_2+\tfrac{1}{2})\omega_{c_2}+(p_{1_z}^2/2m_1\hbar)+(p_{2_z}^2/2m_2\hbar)\pm k\theta/\hbar.$$

To obtain the contribution to the rotation for a transition to a given Landau level in the conduction band we let

$$\sum_k\sum_{k'}\to\sum_{n_1}\sum_{n_2}\int\int\frac{m_1m_2\omega_{c_1}\omega_{c_2}}{16\pi^4\hbar^4}dp_{1_z}dp_{2_z}$$

and perform the indicated integration of Eq. (14) over $\int dp_{1_z}dp_{2_z}$ only. This, together with some algebraic manipulation, leads to the expression

$$\theta_{i(\text{Landau})}=\frac{K_e^{1/2}e^2(m_1m_2)^{3/2}\omega\omega_{c_1}\omega_{c_2}}{256\pi^3m^2c\epsilon\hbar^4}|P_{kk'}|^2\left\{\frac{-8\gamma H\omega}{[\omega^2-(\gamma H)^2]^2}\ln\omega_{12}\tau+\frac{1}{(\omega-\gamma H)^2}\ln[(\omega_{12}-\omega+\gamma H)\tau]\right.$$
$$\left.-\frac{1}{(\omega+\gamma H)^2}\ln[(\omega_{12}+\omega+\gamma H)\tau]-\frac{1}{(\omega+\gamma H)^2}\ln[(\omega_{12}-\omega-\gamma H)\tau]+\frac{1}{(\omega-\gamma H)^2}\ln[(\omega_{12}+\omega-\gamma H)\tau]\right\}, \tag{23}$$

where, in order to make the argument of the logarithms dimensionless, we have anticipated the subsequent introduction of τ and multiplied numerator and denominator of Eq. (14) by τ where appropriate. Here,

$$\omega_{12}=\omega_g+(n_1+\tfrac{1}{2})\omega_{c_1}+(n_2+\tfrac{1}{2})\omega_{c_2}\pm k\theta/\hbar.$$

To obtain the line shape in the vicinity of a given Landau level due to transitions to that level, we proceed as before. We put $(\omega_{12}-\omega\pm\gamma H)\tau\to[(\omega_{12}-\omega\pm\gamma H)\tau+i]$, however, where the terms are of the form $(\omega_{12}+\omega\pm\gamma H)\tau$ we need not introduce the damping since these terms do not give rise to singularities. We seek

$$\theta_{i(\text{Landau})}=\frac{K_e^{1/2}e^2(m_1m_2)^{3/2}\omega_{c_1}\omega_{c_2}}{256\pi^3m^2c\epsilon\hbar^4\omega}|P_{kk'}|^2\left\{\frac{-8\gamma H}{\omega}\ln\omega_{12}\tau+\text{Re}\ln[(\omega_{12}-\omega+\gamma H)\tau+i]\right.$$
$$\left.-\ln[(\omega_{12}+\omega+\gamma H)\tau]-\text{Re}\ln[(\omega_{12}-\omega-\gamma H)\tau+i]+\ln[(\omega_{12}+\omega-\gamma H)\tau]\right\}. \tag{24}$$

Defining $X=(\omega_{12}-\omega)\tau$, $Y=\gamma H\tau$, and $Z=\omega\tau$ we obtain

$$\theta_{i(\text{Landau})}=B\left\{\frac{-16Y}{Z}(\ln Z+\tfrac{1}{8})+\ln[(X+Y)^2+1]-\ln[(X-Y)^2+1]\right\}, \tag{25}$$

where

$$B=\frac{K_e^{1/2}e^2(m_1m_2)^{3/2}\omega_{c_1}\omega_{c_2}}{512\pi^3m^2c\epsilon\hbar^4\omega}|P_{kk'}|^2. \tag{25a}$$

Figures 3(a) and 3(b) are a plot of the expression $\{\ln[(X+Y)^2+1]-\ln[(X-Y)^2+1]\}$. The term $(-16Y/Z)\times(\ln Z+\frac{1}{8})$ is part of the background and need not be considered separately since it will be included in the total background when we integrate over all of the Landau levels.

As for the case of rotation due to direct transitions, one must evaluate the contribution to the indirect rotation due to the background. In the latter case it is necessary to sum over two sets of Landau levels, one each for the conduction and valence bands, since there are no selection rules restricting the indirect transitions. A double integration gives

$$\theta_{i(\text{background})}=\frac{K_e^{1/2}e^2(m_1m_2)^{3/2}\omega}{512\pi^3m^2c\epsilon\hbar^4}|P_{kk'}|^2\bigg\{\frac{-8\gamma H\omega}{[\omega^2-(\gamma H)^2]^2}\omega_g'^2\ln\omega_g'\tau+\frac{1}{(\omega-\gamma H)^2}(\omega_g'-\omega+\gamma H)^2\ln[(\omega_g'-\omega+\gamma H)\tau]$$
$$-\frac{1}{(\omega+\gamma H)^2}(\omega_g'+\omega+\gamma H)^2\ln[(\omega_g'+\omega+\gamma H)\tau]-\frac{1}{(\omega+\gamma H)^2}(\omega_g'-\omega-\gamma H)^2\ln[(\omega_g'-\omega-\gamma H)\tau]$$
$$+\frac{1}{(\omega-\gamma H)^2}(\omega_g'+\omega-\gamma H)^2\ln[(\omega_g'+\omega-\gamma H)\tau]\bigg\}, \quad (26)$$

where $\omega_g'\equiv\omega_g+(k\theta/\hbar)$ to take account of the phonon involved in the transition.

Again in the vicinity of the energy gap we assume $\omega\gg\gamma H$ and let $\omega\tau\to(\omega\tau-i)$ where singularities exist. Letting $X=(\omega_g'-\omega)\tau$, $Y=\gamma H\tau$, and $Z=\omega\tau$ we obtain

$$\theta_{i(\text{background})}=\frac{B}{2\omega_{c1}\omega_{c2}\tau^2}\{-32ZY(\ln Z+0.597)+(X+Y)^2\ln[(X+Y)^2+1]-(X-Y)^2\ln[(X-Y)^2+1]\}. \quad (27)$$

The expression (27) holds subject to the assumption that $X\ll Z$, which defines our region of primary interest. However, if one plots $(X+Y)^2\ln[(X+Y)^2+1]-(X-Y)^2\ln[(X-Y)^2+1]$ in the limit of large X, one sees that it diverges as $4XY(1+2\ln X)$. In this limit, however, one must include the terms that arise from $(\omega_g'+\omega\pm\gamma H)\tau$, namely $(X+2Z+Y)^2\ln(X+2Z+Y)-(X+2Z-Y)^2\ln(X+2Z-Y)$ since here $X\gg Z$. When this is done, the indirect background rotation goes to zero and the mathematical consistency is maintained. Experimentally one would always have $Z\gg X$ and $\omega_{c1}\tau$, $\omega_{c2}\tau\gg1$; the background contribution would then be small compared with the rotation due to the single Landau level.

Indirect Transition—Long-Wavelength Limit

We begin with Eq. (26) since this represents the sum over the contributions from transitions to all Landau levels from all Landau levels. As before, letting $x=(\omega+\gamma H)/\omega_g'$ and $y=(\omega-\gamma H)/\omega_g'$ with $x, y\ll1$, we obtain

$$\theta_i=\frac{K_e^{1/2}e^2(m_1m_2)^{3/2}\omega}{512\pi^3m^2c\epsilon\hbar^4}|P_{kk'}|^2\left\{-\frac{(1+x)^2}{x^2}\ln(1+x)-\frac{(1-x)^2}{x^2}\ln(1-x)+\frac{(1-y)^2}{y^2}\ln(1-y)+\frac{(1+y)^2}{y^2}\ln(1+y)\right\}. \quad (28)$$

Expanding the logarithms and collecting powers of ω_g we finally obtain

$$\theta_i\xrightarrow[\omega\to0]{}\frac{K_e^{1/2}e^2(m_1m_2)^{3/2}}{768\pi^3m^2c\epsilon\hbar^4}|P_{kk'}|^2\frac{\omega^2(\gamma H)}{\omega_g'^2}. \quad (29)$$

VOIGT EFFECT

The Voigt phase shift is given by

$$\delta=\frac{iK_e^{1/2}}{2\epsilon c}(\sigma_{xx}-\sigma_{zz}). \quad (30)$$

Substitution of (3a) into (30) enables us to write

$$\delta=\frac{K_e^{1/2}e^2}{2m^2\hbar\epsilon c}\sum_k\sum_{k'}\frac{1}{\omega_{kk'}}\left\{\frac{P_{kk'}^xP_{k'k}^x}{\omega_{kk'}+\omega}-\frac{P_{kk'}^xP_{k'k}^x}{\omega_{kk'}-\omega}-\left(\frac{P_{kk'}^zP_{k'k}^z}{\omega_{kk'}+\omega}-\frac{P_{kk'}^zP_{k'k}^z}{\omega_{kk'}-\omega}\right)\right\}. \quad (31)$$

Introducing $P^\pm=P^x\pm iP^y$ again, we can obtain

$$|P_{kk'}^+|^2+|P_{kk'}^-|^2=2(P_{kk'}^xP_{k'k}^x+P_{kk'}^yP_{k'k}^y). \quad (32a)$$

One could derive the expressions for the Voigt effect with no restrictions on the matrix elements $P_{kk'}^x$ and $P_{kk'}^y$. However, we will choose for our idealized simple band a cubic crystal symmetry for which $P_{kk'}^xP_{k'k}^x$ and $P_{kk'}^yP_{k'k}^y$ may be interchanged as discussed after Eq. (6); this then enables us to write

$$P_{kk'}^xP_{k'k}^x\to\tfrac{1}{4}\{|P_{kk'}^+|^2+|P_{k'k}^-|^2\}. \quad (32b)$$

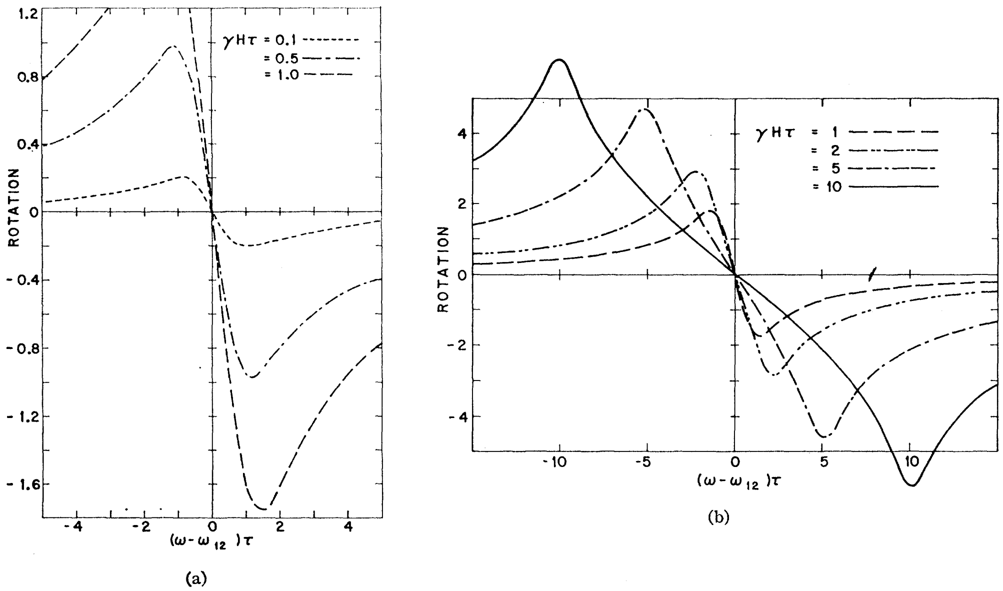


FIG. 3. (a), (b) Plot of the line shapes obtained for the Faraday rotation, indirect transition, between a given pair of Landau levels as a function of frequency for different values of $\gamma H\tau$. The constant B of Eq. (25a) has been normalized to unity.

We then substitute (32b) into (31) to obtain the general expression for the Voigt phase shift.

$$\delta=\frac{K_e^{1/2}e^2\omega}{4m^2\epsilon c\hbar}\sum_k\sum_{k'}\left\{\frac{2|P_{kk'}^{z+}|^2}{\omega_{kk'}^{z+}(\omega_{kk'}^{z+})^2-\omega^2}+\frac{2|P_{kk'}^{z-}|^2}{\omega_{kk'}^{z-}(\omega_{kk'}^{z-})^2-\omega^2}-\frac{|P_{kk'}^{+}|^2}{\omega_{kk'}^{+}(\omega_{kk'}^{+})^2-\omega^2}-\frac{|P_{kk'}^{-}|^2}{\omega_{kk'}^{-}(\omega_{kk'}^{-})^2-\omega^2}\right\}. \tag{33}$$

Note that as in the case for the Faraday rotation we have introduced different denominators for the (+) and (−) transitions since the k and k' are actually dummy variables and the energy denominators must agree with the initial and final states between which the matrix elements are taken.

Equation (33) represents a situation where, corresponding to each "±" pair of excitation energies, there are two "z" states (i.e., a π doublet in the middle of a σ doublet). The frequencies $\omega_{kk'}^{\pm}$ and $\omega_{kk'}^{z\pm}$ are then given by

$$\omega_{kk'}^{\pm}=\omega_{kk'}\pm\gamma_\sigma H\,,\quad \omega_{kk'}^{z\pm}=\omega_{kk'}^{z}\pm\gamma_\pi H\,.$$

In our simple idealized two-band model, we assume, both for analytical tractability and in order to permit comparison with the classical case, that $g_c=g_v$. This then leads to the result that $\gamma_\pi=0$. The π doublet then coalesces to give two "z" states with a single energy which split the σ doublet. We then *define* $\gamma_\sigma\equiv\gamma$ and note that $\omega_{kk'}^{z\pm}\rightarrow\omega_{kk'}^{z}$ and that $|P_{kk'}^{z\pm}|^2\rightarrow|P_{kk'}^{z}|^2$. Equation (33) then reduces to:

$$\delta=\frac{K_e^{1/2}e^2\omega}{4m^2\epsilon c\hbar}\sum_k\sum_{k'}\left\{\frac{4|P_{kk'}^{z}|^2}{\omega_{kk'}^{z}(\omega_{kk'}^{z})^2-\omega^2}-\frac{|P_{kk'}^{+}|^2}{\omega_{kk'}^{+}(\omega_{kk'}^{+})^2-\omega^2}-\frac{|P_{kk'}^{-}|^2}{\omega_{kk'}^{-}(\omega_{kk'}^{-})^2-\omega^2}\right\}. \tag{34}$$

Equation (34) can be used as is as a starting point for evaluating the Voigt phase shift for both the direct and indirect transitions with no further restrictions applied to the matrix elements. However, it lends itself to considerable algebraic simplification to assume a relation between the $|P_{kk'}^{z}|$ and $|P_{kk'}|$ matrix elements in a manner similar to that done for the Faraday effect. This is here obtained by letting that part of the expression within the braces in Eq. (34) be equal to zero for $\omega=0$ and by further assuming that

$$|P_{kk'}|^2/\omega_{kk'}=2(|P_{kk'}^{z}|)^2/(\omega_{kk'}^{z})\,. \tag{35a}$$

Assumption (35a) essentially says that the oscillator strength for the "z" transitions is equal to an oscillator strength, suitably averaged for the "±" transitions. This assumption, together with the substitution $\omega=0$ in Eq. (34), then gives

$$(\omega_{kk'}^{z})^2=\omega_{kk'}^2-(\gamma H)^2\,, \tag{35b}$$

and

$$|P_{kk'}^{z}|^2=\tfrac{1}{2}|P_{kk'}|^2\left(1-\left(\frac{\gamma H}{\omega_{kk'}}\right)^2\right)^{1/2}\rightarrow\tfrac{1}{2}|P_{kk'}|^2\left(1-\tfrac{1}{2}\left(\frac{\gamma H}{\omega_{kk'}}\right)^2\right). \tag{35c}$$

The particular relation (35a) has been chosen in order to make the long-wavelength frequency dependence exhibited by our model agree with that obtained by means of the semiclassical analysis,[8] namely that the effect go as ω^3 for $(\omega_{kk'}-\omega)\gg\gamma H$. Furthermore, as can be seen from Eq. (35c), this choice is consistent with the theoretical work of Burstein *et al.*[17] who show that due to the "band-wave function" contribution, a second-order term in the magnetic field is introduced into the x- and y-matrix elements (or "$\pm$" ones) but not into the z-matrix elements. On the other hand, the line shape behavior in the vicinity of the gap energy will not depend on any higher order correction terms in the magnetic field in the relation between $|P_{kk'}{}^z|$ and $|P_{kk'}|$ since the higher order terms do not give rise to singularities. Equation (35a) together with (11) enables us to express the Voigt phase shift in the form

$$\delta=\frac{-e^2K_e{}^{1/2}\omega}{4m^2\hbar\epsilon c}\sum_k\sum_{k'}\frac{|P_{kk'}|^2}{\omega_{kk'}{}^2}\left\{\frac{\omega_{kk'}{}^+}{(\omega_{kk'}{}^+)^2-\omega^2}+\frac{\omega_{kk'}{}^-}{(\omega_{kk'}{}^-)^2-\omega^2}-\frac{2\omega_{kk'}}{\omega_{kk'}{}^2-[\omega^2+(\gamma H)^2]}\right\}. \tag{36}$$

We now examine the behavior of δ in the limit of small magnetic fields. Expanding Eq. (36) in powers of γH subject to the condition that $\gamma H\ll(\omega_{kk'}-\omega)$, we find that

$$\delta\xrightarrow[\gamma H\to 0]{}\frac{-2e^2K_e{}^{1/2}}{m^2\hbar\epsilon c}\sum_k\sum_{k'}\frac{|P_{kk'}|^2}{\omega_{kk'}}\frac{\omega^3(\gamma H)^2}{(\omega_{kk'}{}^2-\omega^2)^3}. \tag{37}$$

We next expand Eq. (36) in partial fractions to obtain the most general expression for the Voigt phase shift for the model we have chosen [i.e., subject to the conditions on the matrix elements stated in Eq. (35)]. This is

$$\delta=\frac{-e^2K_e{}^{1/2}\omega}{8m^2\hbar\epsilon c}\sum_k\sum_{k'}|P_{kk'}|^2\left\{\frac{-16\omega^2(\gamma H)^2}{[\omega^2-(\gamma H)^2](\omega^4-(\gamma H)^4)\omega_{kk'}}+\frac{1}{(\omega+\gamma H)^2(\omega_{kk'}+\omega+\gamma H)}+\frac{1}{(\omega+\gamma H)^2(\omega_{kk'}-\omega-\gamma H)}\right.$$
$$\left.+\frac{1}{(\omega-\gamma H)^2(\omega_{kk'}+\omega-\gamma H)}+\frac{1}{(\omega-\gamma H)^2(\omega_{kk'}-\omega+\gamma H)}-\frac{2}{\Omega^2(\omega_{kk'}+\Omega)}-\frac{2}{\Omega^2(\omega_{kk'}-\Omega)}\right\}, \tag{38}$$

where we have here put $\Omega^2=\omega^2+(\gamma H)^2$. One also notes from either (34), (36), or (38) that for $H=0$, $\delta=0$, and $\sigma_{xx}=\sigma_{zz}$ as expected.

Direct Transition

To obtain the contribution to the rotation for a direct transition between a given pair of Landau levels we must integrate out p_z from Eq. (38) (where $\omega_{kk'}\to\omega_{kk}=\omega_g+(n+\frac{1}{2})\omega_c+p_z{}^2/2\mu\hbar\equiv\omega_n+p_z{}^2/2\mu\hbar$). This gives

$$\delta_{d\,(\text{Landau})}=\frac{-e^2K_e{}^{1/2}(2\mu)^{3/2}\omega_c\omega}{128\pi m^2\hbar^{5/2}\epsilon c}|P_{kk}|^2\left\{\frac{-16\omega^2(\gamma H)^2}{[\omega^2-(\gamma H)^2][\omega^4-(\gamma H)^4]\omega_n{}^{1/2}}+\frac{1}{(\omega+\gamma H)^2(\omega_n+\omega+\gamma H)^{1/2}}\right.$$
$$+\frac{1}{(\omega+\gamma H)^2(\omega_n-\omega-\gamma H)^{1/2}}+\frac{1}{(\omega-\gamma H)^2(\omega_n+\omega-\gamma H)^{1/2}}+\frac{1}{(\omega-\gamma H)^2(\omega_n-\omega+\gamma H)^{1/2}}$$
$$\left.-\frac{2}{\Omega^2(\omega_n+\Omega)^{1/2}}-\frac{2}{\Omega^2(\omega_n-\Omega)^{1/2}}\right\}. \tag{39}$$

The line shape near a Landau level is obtained as for the Faraday direct, by taking $\omega\gg\gamma H$, picking out the singular terms, and letting $\omega\to\omega-i/\tau$, giving

$$\delta_{d\,(\text{Landau})}=\frac{-K_e{}^{1/2}e^2(2\mu)^{3/2}\tau^{1/2}\omega_c}{128\pi m^2\hbar^{5/2}\epsilon c\omega}|P_{kk}|^2\,\mathrm{Re}\left\{\frac{1}{[(\omega_n-\omega+\gamma H)\tau+i]^{1/2}}+\frac{1}{[(\omega_n-\omega-\gamma H)\tau+i]^{1/2}}-\frac{2}{[(\omega_n-\omega)\tau+i]^{1/2}}\right\}, \tag{40}$$

where for $\omega\gg\gamma H$, $\Omega\to\omega$. Letting $X=(\omega_n-\omega)\tau$ and $Y=\gamma H\tau$,

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[17] E. Burstein, G. S. Picus, R. F. Wallis, and F. Blatt, Phys. Rev. **113**, 15 (1959).

$$\delta_{d(\text{Landau})}=A\left\{\frac{2}{(X^2+1)^{1/2}}[(X^2+1)^{1/2}+X]^{1/2}-\frac{1}{[(X+Y)^2+1]^{1/2}}\{[(X+Y)^2+1]^{1/2}+(X+Y)\}^{1/2}\right.$$
$$\left.-\frac{1}{[(X-Y)^2+1]^{1/2}}\{[(X-Y)^2+1]^{1/2}+(X-Y)\}^{1/2}\right\}, \quad (41)$$

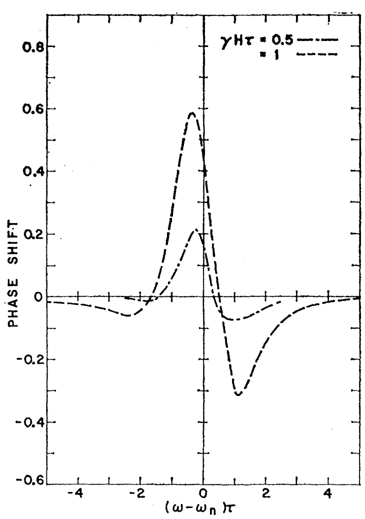
where A is given in Eq. (17a).

The expression in the braces has been plotted in Figs. 4 (a) and 4 (b) with $Y=\gamma H\tau$ as parameter.

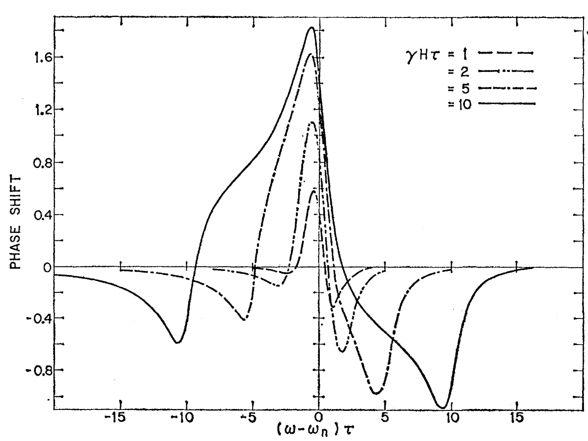
To obtain the background rotation one must sum over n in Eq. (39); this leads to the expression

$$\delta_{d(\text{background})}=\frac{e^2K_e^{1/2}(2\mu)^{3/2}\omega}{64\pi m^2\epsilon c\hbar^{5/2}}|P_{kk}|^2\left\{\frac{-16\omega^2(\gamma H)^2}{[\omega^2-(\gamma H)^2][\omega^4-(\gamma H)^4]}\omega_g^{1/2}\right.$$
$$+\frac{1}{(\omega+\gamma H)^2}(\omega_g+\omega+\gamma H)^{1/2}+\frac{1}{(\omega+\gamma H)^2}(\omega_g-\omega-\gamma H)^{1/2}+\frac{1}{(\omega-\gamma H)^2}(\omega_g+\omega-\gamma H)^{1/2}$$
$$\left.+\frac{1}{(\omega-\gamma H)^2}(\omega_g-\omega+\gamma H)^{1/2}-\frac{2}{\Omega^2}(\omega_g+\Omega)^{1/2}-\frac{2}{\Omega^2}(\omega_g-\Omega)^{1/2}\right\}. \quad (42)$$

Again, in the vicinity of the energy gap we assume $\omega\gg\gamma H$, extract the dominant terms, and let $\omega\rightarrow\omega-i/\tau$ to

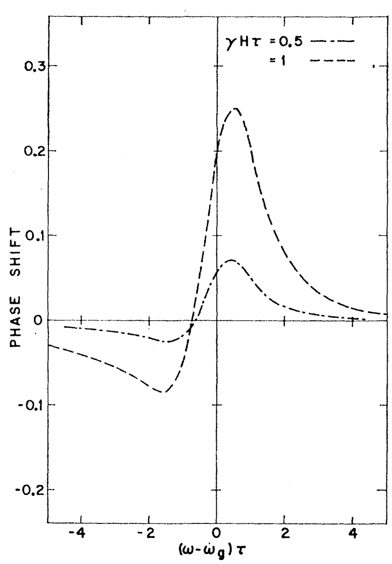


(a)

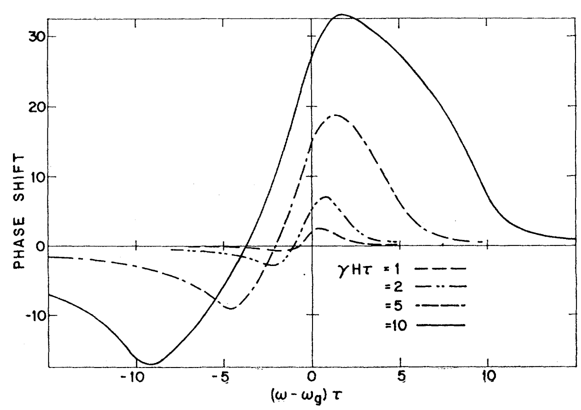


(b)

FIG. 4. (a), (b) Plot of the line shapes for the Voigt phase shift, direct transition, between a given pair of Landau levels as a function of frequency for different values of $\gamma H\tau$. The constant A of Eq. (41) has been normalized to unity.



(a)



(b)

FIG. 5. (a), (b) Contribution to the Voigt phase shift direct transition line shape due to background. The ordinates of the figures must be multiplied by $2/\omega_c\tau$ to permit comparison with Figs. 4 (a), (b).

obtain

$$\delta_{d\,(\text{background})}=-\frac{K_e^{1/2}e^2(2\mu)^{3/2}}{64\pi m^2\epsilon c\hbar^{5/2}\omega\tau^{1/2}}|P_{kk}|^2\,\mathrm{Re}\{2[(\omega_g-\omega)\tau+i]^{1/2}-[(\omega_g-\omega+\gamma H)\tau+i]^{1/2}-[(\omega_g-\omega-\gamma H)\tau+i]^{1/2}\}\,,\qquad(43)$$

which, with the substitution $X=(\omega_g-\omega)\tau$, $Y=\gamma H\tau$, becomes

$$\delta_{d\,(\text{background})}=\frac{2A}{\omega_c\tau}\{\{[(X+Y)^2+1]^{1/2}+(X+Y)\}^{1/2}+\{[(X-Y)^2+1]^{1/2}+(X-Y)\}^{1/2}-2[(X^2+1)^{1/2}+X]^{1/2}\}\,.\qquad(44)$$

We have plotted the expression in the braces in Eq. (44) in Figs. 5(a) and 5(b). A comparison of the ordinates with those in Figs. 4(a) and 4(b) shows that for the order of magnitude of $\omega_c\tau$ that one would expect to obtain either at high magnetic fields or at low temperatures, the background rotation may be neglected compared with the contribution from the single Landau level transition.

Direct Transition—Long-Wavelength Limit

To obtain the behavior of the Voigt phase shift due to the direct transition in the long-wavelength limit, we must sum over the contributions from transitions to all Landau levels [i.e., $\sum_{n=0}^{\infty}\rightarrow\int_0^\infty dn\rightarrow(1/\omega_c)\int_0^\infty dx$, where $x=(n+\frac{1}{2})\omega_c$]. Hence we start with Eq. (42), subject to the condition that $\omega_g\gg\gamma H$, ω. Expansion of Eq. (42) then leads to the result

$$\delta_d\rightarrow-\frac{21e^2K_e^{1/2}(2\mu)^{3/2}}{4096\pi m^2\epsilon c\hbar^{5/2}}|P_{kk}|^2\frac{\omega^3(\gamma H)^2}{\omega_g^{11/2}}\,.\qquad(45)$$

This result shows an ω^3 frequency dependence as has been discussed following Eq. (35).

Indirect Transition

To obtain the contribution to the rotation for an indirect transition to a given Landau level in the conduction band, we must, as for the indirect Faraday case, integrate over all values of p_z, in both the conduction and valence bands. Starting with Eq. (38) and performing the indicated integrations, we obtain

$$\delta_{i\,(\text{Landau})}=\frac{K_e^{1/2}e^2(m_1m_2)^{3/2}\omega_{c1}\omega_{c2}\omega}{256\pi^3m^2\hbar^4\epsilon c}|P_{kk'}|^2\left\{\frac{-16\omega^2(\gamma H)^2}{[\omega^2-(\gamma H)^2][\omega^4-(\gamma H)^4]}\ln\omega_{12}\tau+\frac{1}{(\omega+\gamma H)^2}\ln[(\omega_{12}+\omega+\gamma H)\tau]+\frac{1}{(\omega+\gamma H)^2}\ln[(\omega_{12}-\omega-\gamma H)\tau]+\frac{1}{(\omega-\gamma H)^2}\ln[(\omega_{12}+\omega-\gamma H)\tau]+\frac{1}{(\omega-\gamma H)^2}\ln[(\omega_{12}-\omega+\gamma H)\tau]-\frac{2}{\Omega^2}\ln[(\omega_{12}+\Omega)\tau]-\frac{2}{\Omega^2}\ln[(\omega_{12}-\Omega)\tau]\right\}\,,\qquad(46)$$

where $\omega_{12}=\omega_g'+(n_1+\frac{1}{2})\omega_{c1}+(n_2+\frac{1}{2})\omega_{c2}$, and we have again anticipated the introduction of τ in order to make the argument of the logarithms dimensionless. To obtain the line shapes we make the usual assumptions [$\omega\gg\gamma H$; $\omega\tau\rightarrow(\omega\tau-i)$, where singularities exist] noting that it is necessary to retain all the terms of Eq. (46).

Defining X, Y and Z as before and expanding the logarithms where necessary, since $Z\gg X$, Y we finally obtain

$$\delta_i=B\{-(Y^2/2Z^2)(1+64\ln Z)+\ln[(X+Y)^2+1]+\ln[(X-Y)^2+1]-2\ln[X^2+1]\}\,,\qquad(47)$$

where B is given in Eq. (25a). Figures 6(a) and 6(b) plot the expression $\{\ln[(X+Y)^2+1]+\ln[(X-Y)^2+1]-2\ln[X^2+1]\}$. The quantity $(-Y^2/2Z^2)(1+64\ln Z)$ is part of the background and need not be considered separately.

The background is obtained, as in the case for the Faraday-indirect, by summing over the two sets of Landau levels in the conduction and valence bands, respectively. This leads to the expression

$$\delta_{i\,(\text{background})}=\frac{e^2K_e^{1/2}(m_1m_2)^{3/2}\omega}{512\pi^3m^2\hbar^4\epsilon c}|P_{kk'}|^2\bigg\{\frac{-16\omega^2(\gamma H)^2}{[\omega^2-(\gamma H)^2][\omega^4-(\gamma H)^4]}\omega_g'^2\ln\omega_g'\tau$$

$$+\frac{1}{(\omega+\gamma H)^2}(\omega_g'+\omega+\gamma H)^2\ln[(\omega_g'+\omega+\gamma H)\tau]+\frac{1}{(\omega+\gamma H)^2}(\omega_g'-\omega-\gamma H)^2\ln[(\omega_g'-\omega-\gamma H)\tau]$$

$$+\frac{1}{(\omega-\gamma H)^2}(\omega_g'+\omega-\gamma H)^2\ln[(\omega_g'+\omega-\gamma H)\tau]+\frac{1}{(\omega-\gamma H)^2}(\omega_g'-\omega+\gamma H)^2\ln[(\omega_g'-\omega+\gamma H)\tau]$$

$$-\frac{2}{\Omega^2}(\omega_g'+\Omega)^2\ln[(\omega_g'+\Omega)\tau]-\frac{2}{\Omega^2}(\omega_g'-\Omega)^2\ln[(\omega_g'-\Omega)\tau]\bigg\}. \quad (48)$$

Letting $X=(\omega_g'-\omega)\tau$, $Y=\gamma H\tau$, and $Z=\omega\tau$, we can put the background rotation in the vicinity of a Landau level for the Voigt configuration, indirect transition, into the form

$$\delta_{i\,(\text{background})}\rightarrow\frac{-B}{2\omega_{c_1}\omega_{c_2}\tau^2}\{Y^2(14\ln Z-2\ln 2+3)-(X+Y)^2\ln[(X+Y)^2+1]$$

$$-(X-Y)^2\ln[(X-Y)^2+1]+2X^2\ln(X^2+1)\}. \quad (49)$$

A similar situation applies for the Voigt indirect background as did for the Faraday indirect background. Equation (49) holds in the limit $X\ll Z$ which experimentally is the region in which we are interested. Here $\omega_{c_1}\tau$ and $\omega_{c_2}\tau\gg 1$ and the background contribution is small compared with the rotation due to the single Landau level. However, in the limit of large X, Eq. (49) diverges as $4Y^2\ln X$ and the hitherto neglected terms of Eq. (48) must be included, in which case the indirect background rotation goes to zero.

Indirect Transition—Long-Wavelength Limit

We begin with Eq. (48) since this represents the sum over the contributions to all possible Landau levels in the conduction band from all possible Landau levels in the valence band. Expansion of Eq. (48), subject to the conditions $\omega_g\gg\omega$, γH, gives the result

$$\delta_i\underset{\omega\to 0}{\longrightarrow}-\frac{e^2K_e^{1/2}(m_1m_2)^{3/2}}{1920\pi^3m^2\hbar^4\epsilon c}|P_{kk'}|^2\frac{\omega^3(\gamma H)^2}{\omega_g'^4}, \quad (50)$$

which again show an ω^3 dependence in the long-wavelength limit.

CONDUCTIVITY TENSOR RELATIONS

The conductivity tensor as obtained from first-order time-dependent perturbation theory and given in Eqs. (3a) and (b) can be transformed into some very simple and useful forms through the introduction of left and right circularly polarized waves. We obtain the expressions

$$\sigma^{xx}=\frac{ie^2\omega}{2m^2\hbar}\sum_k\sum_{k'}\bigg\{\frac{|P_{kk'}^+|^2}{\omega_{kk'}^+((\omega_{kk'}^+)^2-\omega^2)}+\frac{|P_{kk'}^-|^2}{\omega_{kk'}^-((\omega_{kk'}^-)^2-\omega^2)}\bigg\}, \quad (51)$$

and

$$\sigma^{xy}=\frac{e^2}{2m^2\hbar}\sum_k\sum_{k'}\bigg\{\frac{|P_{kk'}^+|^2}{(\omega_{kk'}^+)^2-\omega^2}-\frac{|P_{kk'}^-|^2}{(\omega_{kk'}^-)^2-\omega^2}\bigg\}, \quad (52)$$

corresponding to the diagonal and off-diagonal components, respectively. Using the definition

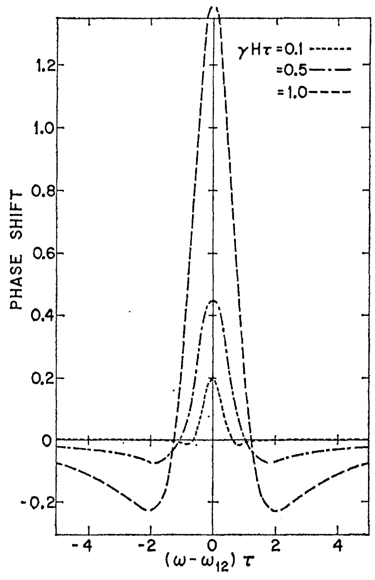
$$\sigma^{\pm}=\sigma^{xx}\mp i\sigma^{xy}, \quad (53)$$

we can also derive expressions for the conductivities associated with the circularly polarized waves; these are given by

$$\sigma^+=\frac{-ie^2}{2m^2\hbar}\sum_k\sum_{k'}\bigg\{\frac{|P_{kk'}^+|^2}{\omega_{kk'}^+(\omega_{kk'}^++\omega)}-\frac{|P_{kk'}^-|^2}{\omega_{kk'}^-(\omega_{kk'}^--\omega)}\bigg\}, \quad (54)$$

and

$$\sigma^-=\frac{ie^2}{2m^2\hbar}\sum_k\sum_{k'}\bigg\{\frac{|P_{kk'}^+|^2}{\omega_{kk'}^+(\omega_{kk'}^+-\omega)}-\frac{|P_{kk'}^-|^2}{\omega_{kk'}^-(\omega_{kk'}^-+\omega)}\bigg\}. \quad (55)$$



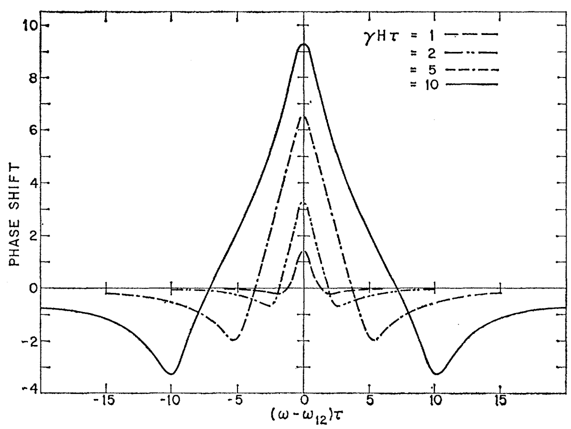


FIG. 6. (a), (b) Plot of the line shapes obtained for the Voigt phase shift, indirect transition, between a given pair of Landau levels as a function of frequency for different values of $\gamma H\tau$. The constant B of Eq. (48) has been normalized to unity.

The interesting point here is that, although each of these conductivities involves either admixtures of the matrix elements and eigenvalues for both left and right circularly polarized waves or implicitly includes transitions between both sets of levels, a still simpler and surprisingly symmetrical form results if we use the conditions obtained from the properties of the Faraday rotation and given by Eq. (11), namely,

$$\frac{|P_{kk'}{}^{+}|^2}{(\omega_{kk'}{}^{+})^2}=\frac{|P_{kk'}{}^{-}|^2}{(\omega_{kk'}{}^{-})^2}=\frac{|P_{kk'}|^2}{\omega_{kk'}{}^2}.$$

We then obtain the following:

$$\sigma^{\pm}=\frac{ie^2\omega}{m^2\hbar}\sum_k\sum_{k'}\frac{|P_{kk'}|^2}{\omega_{kk'}}\left\{\frac{1}{\omega_{kk'}{}^2-(\omega\pm\gamma H)^2}\right\}. \quad (56)$$

This result shows that the form of the oscillator strength for the interband transition given by $|P_{kk'}|^2/\omega_{kk'}$ as postulated by KLN is indeed justified; furthermore, the $\sigma^{\pm}$ conductivities can be interpreted to first order in terms of transforming into a rotating frame in which the magnetic field vanishes while the frequency is changed from ω to $\omega\pm\gamma H$.

DISCUSSION

We have calculated explicit expressions for the Faraday rotation and the Voigt phase shift in the oscillatory region from the off-diagonal and diagonal components of the conductivity tensor which has been obtained in the form of the Kramers-Heisenberg relations. In this paper we have expressed these results in a more general form which is appropriate both to the high-magnetic field limit and/or to the low-temperature case, for which $\gamma H\tau\gg1$. Thus, the expressions we have obtained are valid for such materials as indium antimonide, where at fields of the order of 100 000 G, the results of a theory based on an expansion in powers of the magnetic field would definitely be inappropriate.

The significance of the analysis that we have carried out lies in the behavior of the line shapes as a function of magnetic field and temperature. The line shapes are considerably altered as we go from low values of $\gamma H\tau$, corresponding to room temperature and/or low fields to high values of $\gamma H\tau$ which are obtained at low temperatures and high fields. These results are clearly shown in Figs. 1 (a), (b) and 4 (a), (b) which indicate the direct transition Faraday rotation and Voigt phase shift, respectively.

It was shown by KLN that in the high-temperature case the line shape could be utilized to determine the product of γ and τ since the ratio

$$\delta_{\max}/\theta_{\max}=\text{const}\times\gamma H\tau \quad (57)$$

and that since γ could be obtained from the separation between peaks both γ and τ can be determined. This result can, for the direct transition for instance, be obtained by expanding Eqs. (15) and (39) in powers of γH and then letting $\omega\rightarrow(\omega-i/\tau)$ where singularities exist. The present paper, moreover, shows that in the low-temperature and/or high-field limit, the results are more direct, since now the separation between the maximum and minimum for a given line shape in either the Faraday rotation or the Voigt effect gives γH directly [see Figs. 1 (b) and 4 (b)], and τ can be determined by fitting the line shape.

Finally, it should be pointed out that the results presented in this paper enable one to determine, from the line shape studies, the relative contribution to the dispersion due to transitions between free Landau states and those due to transitions between exciton states. A phenomenological theory based on a simple bound state for the exciton indicates that there is a marked difference between the line shapes for these two types of transitions. At room temperature in germanium the experimentally obtained line shapes appear predominantly to resemble the line shapes calculated in this paper; these correspond to transitions between Landau levels. At low temperatures, where exciton transitions should be favored, there is as yet insufficient data for a proper comparison.

ACKNOWLEDGMENTS

We would like to thank Dr. L. M. Roth for reviewing this manuscript and for suggested changes. We would also like to thank H. S. Bennett, Y. Shapira, Dr. J. Furdyna, and K. J. Button for helpful discussions.

PHYSICAL REVIEW VOLUME 134, NUMBER 1A 6 APRIL 1964

Ultraviolet Optical Properties of Diamond*

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(Received 7 October 1963; revised manuscript received 26 November 1963)

The absolute reflection spectrum of type IIa diamond was measured at room temperature from 4 to 30 eV and analyzed by dispersion techniques to obtain the optical parameters. Structure observed in the dielectric constant near 7, 12, 16, 20, and 24 eV was attributed to interband transitions at critical points in the joint density-of-states function. The new high-energy structure near 16, 20, and 24 eV was assigned to transitions near the L point of the Brillouin zone. Experimental interband transition energies are compared to band theory calculations.

INTRODUCTION

CONSIDERABLE progress has been made recently in our understanding of the detailed physics of tetrahedrally coordinated solids through the comparison of band structure calculations[1] and experimental ultraviolet optical properties.[2] These studies have shown in some detail the similarity in band structure of Ge, Si, and the group III–V compound semiconductors. To a lesser extent this similarity has been shown to extend to certain of the group II–VI compounds. Diamond, the simplest of all tetrahedrally coordinate solids because of its extremely small core composed only the completed $1S$ shell has, however, not received the attention necessary to place it on the same basis as the others of its class in spite of the fact that extensive band calculations were carried out several years ago by Herman[3] using the orthogonalized plane-wave method. Herman's calculations were subsequently shown to be nearly self-consistent by Kleinman and Phillips.[4]

With the exception of a single recent paper by Philipp and Taft,[5] no experimental verification of these calculations has been made for energies greater than that corresponding to the lowest lying transition. In view of the fundamental role of diamond in solid-state physics and the recent interest in its semiconducting form, such verification is desirable.

The present paper reports measurements of the absolute reflection spectrum of a polished, type IIa diamond for the region 4 to 30 eV. Dispersion relation analysis of the reflectivity was used to obtain the complex dielectric constant as well as the optical constants n and k. The improved purity of the diamond specimen employed and increased resolution for energies above 12 eV have lead to significant new high-energy results.

MEASUREMENTS AND ANALYSIS

The absolute reflection spectrum, between 4 and 30 eV of a polished specimen of type IIa diamond[6] was measured at room temperature and at a fixed angle of incidence of 20°. The absolute accuracy of the reflectivity was determined mainly by the experimental scatter which amounted at most to 5% in the region between 12 and 20 eV. Both below and above this region the absolute accuracy of the data was about 3% and the relative accuracy somewhat better. Averaging of several independent measurements allowed structure amounting to 1 or 2% to be determined.

A Jarrell-Ash one-half meter Seya monochromator was used with a bandpass ranging from 3 to 6 Å giving an energy resolution between 0.005 and 0.1 eV depending on the spectral region involved. In the important region between 12 and 20 eV the maximum spacing between source emission lines was 0.7 eV and was more

* Work supported by the National Aeronautics and Space Administration.
† Currently at Goddard Space Flight Center under a NASA Post-Doctoral Fellowship (on leave from the Physics Department, University of California, Santa Barbara, California).
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[6] The type-IIa diamond, a rectangular parallelipiped measuring 1.09×2.98×4.96 mm and weighing 0.28 carat was graciously supplied by Dr. F. A. Raal of the Diamond Research Laboratory, Crown Mines, Johannesburg, South Africa.