Magnetic Corrections to the Boltzmann Transport Equation

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The usual derivations of the Boltzmann transport equation suffer from a number of weaknesses. Using a simple model, this paper attempts to treat on as rigorous a basis as possible the influence of a magnetic field on the transport equation. The approach presented here establishes the relationship between the classical Boltzmann equation and the corresponding quantum-mechanical formalism. It is shown how the exact gauge-dependent Liouville equation, determining the density matrix, can be transformed into a completely gauge-independent equation satisfied by a new density matrix. This equation is solved for a model consisting of noninteracting free electrons being elastically scattered by randomly placed scattering centers. The new density matrix is developed in ascending powers of the strength of the scattering potential. In carrying out this development, the product of the cyclotron frequency and the collision relaxation time is assumed to be of order unity. In this case the familiar Boltzmann transport equation in the presence of a magnetic field represents an approximation valid in the limiting cases of very weak or very dilute scatterers. The corresponding velocity operator is shown to be the usual gauge-independent expression, just the ordinary free-particle momentum operator divided by the mass. Higher order corrections to the transport equation are found, some of which involve the magnetic field.

I. INTRODUCTION

THE various effects of a magnetic field on the transport properties of a metal or plasma provide us with some of our most useful information about these substances. For this reason, the dependence of the transport equation on the magnetic field should be established as firmly as possible. In this paper we shall be concerned with treating on as rigorous a basis as possible the influence of a magnetic field on the transport equation using a somewhat simplified model. The approach presented here establishes the relationship between the classical Boltzmann equation and the corresponding gauge-dependent quantum-mechanical formalism. It also provides a consistent method of obtaining the magnetic corrections to the ordinary Boltzmann transport equation to all orders.

A system containing a large number of interacting particles is ordinarily treated by means of a transport equation.[1] This is an equation for a distribution function which describes the probability of a particle being in any given state of motion specified by a suitable set of variables. This equation is usually determined by requiring that in the steady state the total time rate of change of the distribution function vanish. Two essentially distinct processes contribute to changes of distribution function with time, namely, acceleration of charged particles by electric and magnetic fields and collisions between the various particles. The transport equation can, therefore, be written as the sum of two partial derivatives,

$$(\partial f/\partial t)_{\text{Fields}}+(\partial f/\partial t)_{\text{Collisions}}=0, \qquad (1.1)$$

where the first term is the so-called "drift" term and the second is the collision term.

Classical arguments have been used to obtain explicit expressions for the two terms contained in Eq. (1.1). The resulting equation is the well-known Boltzmann transport equation in which the electric and magnetic fields enter in the form of the Lorentz force.[1] The extension of Eq. (1.1) to include quantum-mechanical processes was first considered by Jones and Zener.[2] They examined the action of uniform electric and magnetic fields on the motion of an electron using quantum mechanics and showed that for times small compared to the cyclotron period, the field term in Eq. (1.1) could be written

$$\left(\frac{\partial f}{\partial t}\right)_{\text{Fields}}=e\left\{\mathbf{E}+\frac{1}{c}\frac{\mathbf{k}}{m}\times\mathbf{B}\right\}\cdot\nabla_k f, \qquad (1.2)$$

where $\mathbf{k}$ is related to the velocity and momentum by $\mathbf{k}=m\mathbf{v}=\mathbf{p}$. (Units are chosen such that $h=1$.) In obtaining the result (1.2) Jones and Zener started with the appropriate Schrödinger equation for electrons in the presence of uniform electric and magnetic fields. The vector potential then appears explicitly in the Hamiltonian and both the distribution function and the corresponding velocity operator can be expected to depend on the choice of gauge. This is actually the case; it was recognized that a degree of arbitrariness exists in the definition of the velocity operator for a system subjected to an externally applied magnetic field. The choice of gauge, however, cannot affect in any way the physical properties of the system although the equations determining the motion of the charged particles are clearly different for different choices of gauge. It should, therefore, be possible to carry out the formulation with an arbitrary choice of gauge and arrive finally at a set of equations in which all arbitrariness has disappeared. There appeared at the time to be no simple way of accomplishing this and it was easier to specify an absolute gauge system by simply requiring the mean value of the vector potential to be zero. This was sufficient to

[1] S. Chapman and T. Cowling, *The Mathematical Theory of Non-Uniform Gases*, 3rd ed. (Cambridge University Press, Cambridge, England, 1958), Chap. 3.

[2] H. Jones and C. Zener, Proc. Roy. Soc. (London) **A144**, 101 (1934).

arrive at the expression (1.2) for the field term and at the same time regain the usual expression $\mathbf{p}/m$ for the velocity even in the presence of a magnetic field.

Even so Eq. (1.1) itself cannot properly be taken as a basis for developing a quantum theory of transport for it is well known that a quantum-mechanical calculation of the average of a physical quantity requires in general not only the probabilities of the different states being occupied (the diagonal elements of the density matrix) but the entire density matrix. Here the procedures of Kohn and Luttinger[3] will be followed to obtain in a consistent way the magnetic corrections to the transport equation, which are at the same time independent of the gauge. Since the interaction with the magnetic field is our principal concern, a very simple model will be considered; it will consist of noninteracting free electrons elastically scattered by randomly distributed scattering centers in the presence of externally applied electric and magnetic fields. This is a reasonable description of either a plasma or a metal under certain conditions. It is at once apparent, however, that the diagonal elements of the density matrix for the system in the presence of a magnetic field cannot correspond directly to the classical distribution function satisfying the ordinary Boltzmann transport equation since the equation determining the density operator must depend explicitly on the gauge. Moreover the matrix equations obtained by applying the Kohn-Luttinger procedure directly contain matrix elements of the vector potential itself and this leads directly to difficulty in attempting to solve them. In order to avoid this difficulty and to ensure a description that is finally gauge-independent, the vector potential is eliminated from the operator equations initially. In Sec. II the equation for the exact density operator is shown to be equivalent to a new gauge-independent operator equation, whose solution does in fact correspond to the ordinary classical distribution function. The velocity operator corresponding to the new, or transformed, density operator turns out to be just the ordinary velocity operator one finds in the absence of a magnetic field. The methods of Kohn and Luttinger can be applied at once to the transformed operator equation. This is done in Sec. III to obtain the lowest order equation satisfied by the diagonal elements of the transformed density matrix. This equation turns out to be the ordinary Boltzmann transport equation in the presence of a magnetic field. In Sec. IV the first correction to the transport equation involving the magnetic field is found by extending the work of Sec. III. The new term in the transport equation depends on the magnetic field and the scattering potential. Its contribution to the current density will be treated in a later publication. In the Appendix a proof of an operator theorem used in Sec. II is presented.

[3] W. Kohn and J. M. Luttinger, Phys. Rev. **108**, 590 (1957), hereafter referred to as K-L.

II. GAUGE-INDEPENDENT FORMULATION OF TRANSPORT THEORY

Consider a collection of electrons so dilute that their interaction with each other and the effect of Fermi-Dirac statistics on their behavior can be neglected. Then every electron may be treated as completely independent of all others and one has essentially to deal with a single-electron problem. The electrons are treated as completely free except for their interaction with a set of randomly distributed scattering centers and the externally applied electric and magnetic fields. The scattering centers might be thought of as either more slowly moving ions in a plasma or impurities in a metal. The total Hamiltonian for each electron moving in such a system may be written

$$H_T=H(\mathbf{p}-(e/c)\mathbf{A})+H'(\mathbf{r})+H_E(\mathbf{r}), \tag{2.1}$$

where $H(\mathbf{p}-(e/c)\mathbf{A})$ is the Hamiltonian of a free electron in the presence of a magnetic field $\mathbf{B}$, $H'(\mathbf{r})$ is the scattering potential interaction, assumed to depend only on the coordinates, and $H_E(\mathbf{r})$ is the interaction with the externally applied electric field. The various terms in Eq. (2.1) are given by

$$H(\mathbf{p}-(e/c)\mathbf{A})=(1/2m)\left(\mathbf{p}-\frac{e}{c}\mathbf{A}(\mathbf{r})\right)^2, \tag{2.2}$$

$$H'(\mathbf{r})=\sum_{i=1}^{N}\phi(\mathbf{r}-\mathbf{r}_i)=\lambda V, \tag{2.3}$$

$$H_E(\mathbf{r})=-e\mathbf{E}\cdot\mathbf{r}. \tag{2.4}$$

In (2.2), $\mathbf{A}(\mathbf{r})$ is the vector potential and e is the charge of the electron. In (2.3), λ is some dimensionless measure of the strength of the interaction of the scattering center with the electron, $\phi(\mathbf{r})$ is the interaction energy with a single scatterer, and $\mathbf{r}_i$ are the locations of the N scatterers. In (2.4) $\mathbf{E}$ is the externally applied electric field.

Consider a collection of ν electrons moving under the action of the same Hamiltonian H_T and introduce the exact density operator ρ_T for this collection.[4] Denote the time-dependent wave functions of the electrons by $\Psi^i(\mathbf{r},t)$ and expand them in a complete set of time-independent functions $\psi_l(\mathbf{r})$;

$$\Psi^i(\mathbf{r},t)=\textstyle\sum_l a_l^i(t)\psi_l(\mathbf{r}).$$

Then the Hermitian operator $\rho_T(\mathbf{p},\mathbf{r},t)$ with the matrix elements

$$[\rho_T]_{ll'}=\frac{1}{\nu}\sum_{i=1}^{\nu}a_l^i(t)a_{l'}^{i*}(t)$$

in the $\psi_l(\mathbf{r})$ representation is the exact density operator. The expectation value of any observable quantity

[4] R. C. Tolman, *Principles of Statistical Mechanics* (Oxford University Press, New York, 1930), p. 327.

represented by the operator $M(t)$ is then given by

$$\bar{M}(t)=\mathrm{Tr}\{\rho_T(t)M(t)\}, \tag{2.5}$$

where Tr means the trace or diagonal sum of the corresponding matrix. The time development of $\rho_T(t)$ is determined by the one-electron Liouville equation,

$$i\partial\rho_T/\partial t=[H_T,\rho_T] \tag{2.6}$$

for the Hamiltonian (2.1). The diagonal elements $[\rho_T]_{ll}$ give the probability of finding an electron in the state $\psi_l(r)$. The sum of these probabilities is unity for a wave function normalized to unity. Once ρ_T is known the observed value of any operator representing a physical quantity can be found from Eq. (2.5).

In the absence of an electric field the system can exist in a state of equilibrium. The exact equilibrium density operator ρ must then satisfy the equation

$$i\partial\rho/\partial t=[H(\mathbf{p}-(e/c)\mathbf{A})+H'(\mathbf{r}),\rho]. \tag{2.7}$$

While any function $H(\mathbf{p}-(e/c)\mathbf{A})+H'(\mathbf{r})$ will satisfy this equation, it is known from the theory of statistical mechanics that the exact form of the function must be the usual Maxwell-Boltzmann distribution,

$$\rho=K\exp\{-\beta[H(\mathbf{p}-(e/c)\mathbf{A})+H'(\mathbf{r})]\}, \tag{2.8}$$

where $\beta=1/kT$. The normalization constant K must be determined from the relation

$$K^{-1}=\mathrm{Tr}\{\exp\{-\beta[H(\mathbf{p}-(e/c)\mathbf{A})+H'(\mathbf{r})]\}\} \tag{2.9}$$

and will in general depend on both the magnetic field and the scattering potential.

Now assume that ρ_T satisfying Eq. (2.6) can be written in the form

$$\rho_T=\rho_T(\mathbf{p}-(e/c)\mathbf{A},\mathbf{r},\mathbf{B},t). \tag{2.10}$$

This implies that ρ_T depends not only on the vector potential $\mathbf{A}$ through the operator $\mathbf{p}-(e/c)\mathbf{A}$, but on the gauge-invariant magnetic field $\mathbf{B}$ as well. For the purpose of obtaining a gauge-independent theory of transport we make use of (2.10) to write ρ_T as a Fourier integral

$$\rho_T(\mathbf{p}-(e/c)\mathbf{A},\mathbf{r},\mathbf{B},t)=\int e^{i\mathbf{P}\cdot\xi}R(\xi,\mathbf{r},\mathbf{B},t)d\xi, \tag{2.11}$$

where $\mathbf{P}\equiv(\mathbf{p}-(e/c)\mathbf{A})$. This in turn defines the function $R(\xi,\mathbf{r},\mathbf{B},t)$, which is assumed to depend in no way on the magnetic field gauge. The assumption (2.10) or (2.11) will be justified by showing that the results finally obtained are consistent with it. Now replace ρ_T in Eq. (2.6) by the integral expression (2.11). This gives

$$i\int e^{i\mathbf{P}\cdot\xi}\frac{\partial R}{\partial t}d\xi=\left[H'(\mathbf{r})+H_E(\mathbf{r}),\int e^{i\mathbf{P}\cdot\xi}Rd\xi\right]+\frac{1}{2m}\left[\mathbf{P}^2,\int e^{i\mathbf{P}\cdot\xi}Rd\xi\right]. \tag{2.12}$$

We now wish to rewrite the two commutators on the right-hand side so that the vector potential appears only in an exponential-operator factor to the extreme left in the equivalent expressions. The term on the left-hand side is already in this form since $R(\xi,\mathbf{r},\mathbf{B},t)$ has been assumed to be independent of the gauge. This procedure will allow us to find a gauge-independent equation satisfied by R.

Since the two potential functions depend only on the coordinates, one can easily show that

$$\left[H'+H_E,\int e^{i\mathbf{P}\cdot\xi}Rd\xi\right]=\int e^{i\mathbf{P}\cdot\xi}\times\{e^{-i\mathbf{p}\cdot\xi}(H'+H_E)e^{i\mathbf{p}\cdot\xi}-(H'+H_E)\}Rd\xi \tag{2.13}$$

by using the operator expansion

$$e^{iA}Be^{-iA}=\sum_{n=0}^{\infty}\frac{(i)^n}{n!}[A^{[n]},B], \tag{2.14}$$

in which $[A^{[n]},B]$ is defined to be the n-fold commutator of operator A with operator B, i.e., $[A,[A\cdots],B]$ with n factors of A, and $[A^{[0]}B]\equiv B$. Equation (2.13) allows the first commutator on the right-hand side of (2.12) to be written in the required form.

The second commutator in (2.12) requires more work in order to reduce it to the proper form. First expand it in the following way:

$$\left[\mathbf{P}^2,\int e^{i\mathbf{P}\cdot\xi}Rd\xi\right]=\int e^{i\mathbf{P}\cdot\xi}[\mathbf{P}^2,R]d\xi+\int[\mathbf{P}^2,e^{i\mathbf{P}\cdot\xi}]Rd\xi. \tag{2.15}$$

The first integral on the right can in turn be expanded and written

$$\int e^{i\mathbf{P}\cdot\xi}[\mathbf{P}^2,R]d\xi=\sum_\alpha\int e^{i\mathbf{P}\cdot\xi}\left\{2P_\alpha[P_\alpha,R]+\frac{\partial^2R}{\partial x_\alpha{}^2}\right\}d\xi \tag{2.16}$$

by successive application of the commutator relations. With no loss of generality the magnetic field is chosen in the z direction. The commutators $[P_\alpha,P_\beta]$ are always constants and equal in this case to

$$\begin{aligned}&[P_x,P_y]=i(e/c)B_z,\\&[P_x,P_z]=[P_y,P_z]=0,\end{aligned} \tag{2.17}$$

for any gauge. Further notice that if A and B are any two operators which both commute with their commutator $[A,B]$ then[5]

$$e^{A+B}=e^Ae^Be^{-1/2[A,B]}. \tag{2.18}$$

[5] E. Merzbacher, *Quantum Mechanics* (John Wiley & Sons, Inc., New York, 1961), p. 162.

It is evident from (2.17) that P_α and P_β are two such operators. Now examine the terms contained in the commutator on the right-hand side of Eq. (2.16). With the help of Eqs. (2.17) and (2.18) the first term in the sum over α can be written

$$\int e^{i\mathbf{P}\cdot\xi}P_x[P_x,R]d\xi=i\int e^{i\mathbf{P}\cdot\xi}\left[p_x,\frac{\partial R}{\partial\xi_x}\right]d\xi + \frac{eB}{2c}\int e^{i\mathbf{P}\cdot\xi}\xi_y[p_x,R]d\xi \quad (2.19)$$

after performing a partial integration with respect to ξ_x and assuming that

$$\lim_{\xi_\alpha\to\infty} R(\xi,\mathbf{r},\mathbf{B},t)=0. \quad (2.20)$$

This last condition is certainly well satisfied in all cases of phyaical interest. In a similar way it can be shown that

$$\int e^{i\mathbf{P}\cdot\xi}P_y[P_y,R]d\xi=i\int e^{i\mathbf{P}\cdot\xi}\left[p_y,\frac{\partial R}{\partial\xi_y}\right]d\xi - \frac{eB}{2c}\int e^{i\mathbf{P}\cdot\xi}\xi_x[p_y,R]d\xi, \quad (2.21)$$

$$\int e^{i\mathbf{P}\cdot\xi}P_z[P_z,R]d\xi=i\int e^{i\mathbf{P}\cdot\xi}\left[p_z,\frac{\partial R}{\partial\xi_z}\right]d\xi. \quad (2.22)$$

Finally the last commutator on the right-hand side of Eq. (2.15) must be reduced to proper form. It can be rewritten

$$\int[\mathbf{P}^2,e^{i\mathbf{P}\cdot\xi}]Rd\xi=\frac{2ieB}{c}\int e^{i\mathbf{P}\cdot\xi}\left\{\xi_x\frac{\partial R}{\partial\xi_y}-\xi_y\frac{\partial R}{\partial\xi_x}\right\}d\xi \quad (2.23)$$

by using the commutator relations

$$\begin{aligned}[P_x,e^{i\mathbf{P}\cdot\xi}]&=-(eB/c)e^{i\mathbf{P}\cdot\xi}\xi_y,\\ [P_y,e^{i\mathbf{P}\cdot\xi}]&=(eB/c)e^{i\mathbf{P}\cdot\xi}\xi_x,\\ [P_z,e^{i\mathbf{P}\cdot\xi}]&=0,\end{aligned} \quad (2.24)$$

and finally performing an appropriate partial integration with respect to ξ using Eq. (2.20). The commutators (2.24) can easily be derived by combining Eqs. (2.14) and (2.17).

Placing these results in Eq. (2.12) shows that it is equivalent to the equation

$$\int e^{i\mathbf{P}\cdot\xi}\Big\{-i\frac{\partial R}{\partial t}+e^{-i\mathbf{p}\cdot\xi}(H'+H_E)e^{i\mathbf{p}\cdot\xi}R-R(H'+H_E) +\frac{i}{m}\sum_\alpha\left[p_\alpha,\frac{\partial R}{\partial\xi_\alpha}\right]+\frac{\omega_0}{2}(\xi_y[p_x,R]-\xi_x[p_y,R]) +\frac{1}{2m}\sum_\alpha\frac{\partial^2 R}{\partial x_\alpha^2}+i\omega_0\left(\xi_x\frac{\partial R}{\partial\xi_y}-\xi_y\frac{\partial R}{\partial\xi_x}\right)\Big\}d\xi=0, \quad (2.25)$$

where ω_0 is the cyclotron frequency. This now has the proper form; all of the gauge dependence has been relegated to the single exponential operator factor at the left. The remaining part of the integrand does not depend on the gauge in any way but only on the coordinates. The apparent asymmetry results solely from the special choice of magnetic field direction. If $\mathbf{P}$ were not an operator (2.25) would imply at once that the remaining factor in the integrand must vanish. The theorem is still true even when $\mathbf{P}$ is an operator; this is shown in the Appendix. Hence R must satisfy the gauge-independent equation obtained by setting the expression within the curly bracket in (2.25) equal to zero.

In order to find an equation satisfied by a gauge-independent density operator we define the new function

$$\tilde\rho_T(\mathbf{p},\mathbf{r},\mathbf{B},t)=\int e^{i\mathbf{p}\cdot\xi}R(\xi,\mathbf{r},\mathbf{B},t)d\xi. \quad (2.26)$$

Notice that according to (2.11) $\tilde\rho_T$ is just equal to ρ_T with the vector potential set equal to zero. Now multiply our equation for R, obtained from Eq. (2.25), by the factor $e^{i\mathbf{p}\cdot\xi}$ and integrate over ξ. The new function defined by Eq. (2.26) is then found to satisfy the equation.

$$i\frac{\partial\tilde\rho_T}{\partial t}=[H'+H_E,\tilde\rho_T]+\frac{1}{2m}\sum_\alpha\frac{\partial^2\tilde\rho_T}{\partial x_\alpha^2}+\frac{i}{m}\int e^{i\mathbf{p}\cdot\xi} \times\sum_\alpha\left[p_\alpha,\frac{\partial R}{\partial\xi_\alpha}\right]d\xi+\tfrac{1}{2}\omega_0\int e^{i\mathbf{p}\cdot\xi}\{\xi_y[p_x,R]-\xi_x[p_y,R]\}d\xi +i\omega_0\int e^{i\mathbf{p}\cdot\xi}\left\{\xi_x\frac{\partial R}{\partial\xi_y}-\xi_y\frac{\partial R}{\partial\xi_x}\right\}d\xi. \quad (2.27)$$

After considerable manipulation involving some partial integrations we find that $\tilde\rho_T$ must satisfy the gauge-independent equation

$$i\frac{\partial\tilde\rho_T}{\partial t}=[H_0+H'(\mathbf{r})+H_E(\mathbf{r}),\tilde\rho_T] -\frac{e}{2mc}\sum_\alpha\{(\mathbf{p}\times\mathbf{B})_\alpha[x_\alpha,\tilde\rho_T]+[x_\alpha,\tilde\rho_T](\mathbf{p}\times\mathbf{B})_\alpha\}, \quad (2.28)$$

where

$$H_0=(1/2m)\mathbf{p}^2 \quad (2.29)$$

is just the free-electron Hamiltonian.

In order to use Eq. (2.28) as the basis of a gauge-independent transport theory the correct velocity operator corresponding to it must first be found. It will turn out to be also gauge-independent. To find such an operator we begin with the exact expression for the average current density given, according to formula (2.5), by

$$\langle\mathbf{J}\rangle_{\mathrm{av}}=en_0\,\mathrm{Tr}\{\mathbf{v}\rho_T\}, \quad (2.30)$$

where n_0 is the electron density and $\mathbf{v}$ is the exact velocity operator defined as the total time rate of change

of the spatial coordinate. For the Hamiltonian (2.1) the exact velocity operator must be[6]

$$\begin{aligned}\mathbf{v}&=i[H(\mathbf{p}-(e/c)\mathbf{A})+H'(\mathbf{r})+H_E(\mathbf{r}),\mathbf{r}]\\&=\frac{1}{m}(\mathbf{p}-(e/c)\mathbf{A}(\mathbf{r}))=(1/m)\mathbf{P}\end{aligned}\tag{2.31}$$

and is seen to depend explicitly on the magnetic field through the vector potential. Inserting this in Eq. (2.30) along with the integral expression (2.11) for the exact density operator gives for the current density

$$\langle\mathbf{J}\rangle_{\mathrm{av}}=\frac{en_0}{m}\,\mathrm{Tr}\left\{\mathbf{P}\int e^{i\mathbf{P}\cdot\xi}R(\xi,\mathbf{r},\mathbf{B},t)d\xi\right\}.\tag{2.32}$$

The current density cannot depend in any way on the magnetic field gauge. Hence it is permissible to choose any gauge that might be convenient for the purpose of calculating the trace. The operation of taking the trace in Eq. (2.32) must be sufficient to render the result totally gauge independent. This will now be shown explicitly by writing Eq. (2.32) in terms of the transformed gauge-independent density operator. The form of the corresponding velocity operator and the fact that it too is gauge-independent will then be apparent.

With the help of formulas (2.17) and (2.18) the x component of the average current density can be written

$$\begin{aligned}\langle J_x\rangle_{\mathrm{av}}&=\frac{en_0}{im}\,\mathrm{Tr}\left\{\int\left(\frac{\partial}{\partial\xi_x}e^{iP_x\xi_x}\right)e^{i(P_y\xi_y+P_z\xi_z)}\right.\\&\qquad\left.\times e^{(1/2)im\omega_0\xi_x\xi_y}Rd\xi\right\}\\&=\frac{ien_0}{m}\,\mathrm{Tr}\left\{\int e^{iP_x\xi_x}e^{i(P_y\xi_y+P_z\xi_z)}e^{(1/2)im\omega_0\xi_x\xi_y}\right.\\&\qquad\left.\times\left[\tfrac{1}{2}im\omega_0\xi_yR+\frac{\partial R}{\partial\xi_x}\right]d\xi\right\}\end{aligned}\tag{2.33}$$

after performing a partial integration with respect to ξ_x. To calculate the trace we choose a Landau gauge, $\mathbf{A}(\mathbf{r})=(-yB,0,0)$, and a plane-wave representation. (See Sec. III.) In this case the x component of the average current density becomes

$$\begin{aligned}\langle J_x\rangle_{\mathrm{av}}&=\frac{ien_0}{m}\sum_k\sum_{k'}\sum_{k''}\int e^{ik_x'\xi_x}\delta_{k_xk_x'}\\&\quad\times\delta_{k_y+\omega_0m\xi_x\delta_{k_zk_z'}}e^{i(k_y''\xi_y+k_z''\xi_z)}\\&\quad\times\delta_{k''k}e^{\frac{1}{2}im\omega_0\xi_x\xi_y}\left[\tfrac{1}{2}im\omega_0\xi_yR_{k''k}+\frac{\partial R_{k''k}}{\partial\xi_x}\right]d\xi\\&=\frac{ien_0\Omega}{m}\left[\frac{\partial R_{kk}}{\partial\xi_x}\right]_{\xi=0},\end{aligned}\tag{2.34}$$

where the matrix of R is given by

$$R_{kk'}=\frac{1}{\Omega}\int e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}}R(\xi,\mathbf{r},\mathbf{B},t)d\mathbf{r}.\tag{2.35}$$

The subscript $\xi=0$ in the final expression means that the bracket is to be evaluated at the origin. Then according to (2.35) $\langle J\rangle_{\mathrm{av}}$ no longer depends on the wave vector.

The expression (2.34) for the current density can be put in a more revealing form by making use of the definition of the delta function expressed as a sum:

$$\textstyle\sum_k e^{i\mathbf{k}\cdot\mathbf{r}}=\Omega\delta(\mathbf{r}).\tag{2.36}$$

Using this Eq. (2.34) can be rewritten

$$\begin{aligned}\langle J_x\rangle_{\mathrm{av}}&=\frac{ien_0}{m}\sum_k\int e^{i\mathbf{k}\cdot\xi}\frac{\partial R_{kk}}{\partial\xi_x}d\xi,\\&=\frac{en_0}{m}\sum_k\int k_xe^{i\mathbf{k}\cdot\xi}R_{kk}d\xi,\\&=(en_0/m)\,\mathrm{Tr}\{p_x\tilde{\rho}_T(\mathbf{p},\mathbf{r},\mathbf{B},t)\},\end{aligned}\tag{2.37}$$

by performing one partial integration and using the definition of the transformed gauge-independent density operator given by Eq. (2.26). The remaining components of the average current density can be brought into exactly the same form as (2.37). The resulting expression for the current density is completely equivalent to (2.30) but is expressed in manifestly gauge-independent operators. If $\tilde{\rho}_T$ satisfying Eq. (2.28) is looked upon as the correct density operator, then it is clear from our last results that the free-particle momentum operator divided by the mass is the corresponding velocity operator.

In order to use Eq. (2.28) as the basis of a gauge-independent transport theory, the trace of the transformed density operator $\tilde{\rho}_T(\mathbf{p},\mathbf{r},\mathbf{B},t)$ must be known. This is readily determined by the same methods already used in finding the gauge-independent velocity operator. The trace of the exact density operator has been set equal to unity according to Eq. (2.9). Making use of Eq. (2.26) the trace of ρ_T can be rewritten

$$\begin{aligned}1&=\mathrm{Tr}\left\{\int e^{i\mathbf{P}\cdot\xi}R(\xi,\mathbf{r},\mathbf{B},t)d\xi\right\}\\&=\sum_k\sum_{k'}\sum_{k''}\int e^{ik_x'\xi_x}\delta_{k_xk_x'}e^{\frac{1}{2}im\omega_0\xi_x\xi_y}\delta_{k_yk_y'+m\omega_0\xi_x}\\&\qquad\times\delta_{k_zk_z'}e^{i(k_y''\xi_y+k_z''\xi_z)}\delta_{k'k''}R_{\mathbf{k}''\mathbf{k}}d\xi\\&=\Omega[R_{kk}]_{\xi=0},\end{aligned}\tag{2.38}$$

[6] See, for example, L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), p. 132.

where the matrix of R, given by (2.35), is to be evaluated at the origin. This expression for the trace can also be written

$$1=\Omega\int\delta(\xi)R_{kk}d\xi$$

$$=\sum_k\sum_{k'}\int e^{i\mathbf{k}\cdot\xi}R_{kk'}\delta_{kk'}d\xi$$

$$=\mathrm{Tr}\tilde{\rho}_T(\mathbf{p},\mathbf{r},\mathbf{B},t) \tag{2.39}$$

by using Eq. (2.36) and the definition of $\tilde{\rho}_T$ given by (2.26). Thus the trace of the gauge-independent density operator is the same as that of the exact density operator ρ_T. Equation (2.39) can then be used to determine the normalization of $\tilde{\rho}_T$.

III. BOLTZMANN TRANSPORT EQUATION

The problem of handling the gauge-dependent operator Eq. (2.6) has been reduced to the equivalent problem of treating the gauge-independent Eq. (2.28). It will now be shown that it is $\tilde{\rho}_T$ and not ρ_T that actually corresponds to the ordinary classical distribution function. This is accomplished by developing the new density matrix in ascending powers of the strength of the scattering potential according to a technique used by Kohn and Luttinger.[3] In order to obtain the familiar Boltzmann transport equation, which represents the lowest order approximation, it is necessary to assume that $\omega_0\tau_r\approx 1$ where ω_0 is the cyclotron frequency and τ_r is of the order of the collision relaxation time. Higher order corrections to the Boltzmann equation exist, some of which involve the magnetic field explicitly. These will be dealt with in the last section; here we consider only the lowest order contribution.

Consider Eq. (2.28) in the following way. Initially the system is in contact with a heat reservoir and the system is assumed to have reached thermodynamic equilibrium in the presence of a magnetic field. There is no electric field present. Contact is then broken with the heat reservoir and the electric field is very slowly turned on. It is convenient to turn the electric field on according to the formula

$$\mathbf{E}=\mathbf{E}^0e^{st} \tag{3.1}$$

so that the electric field is zero at $t=-\infty$ and reaches its full value $\mathbf{E}^0$ at time zero. It will be shown that the results do not depend on the frequency parameter s as long as it is chosen reasonably. The collection of electrons is now described by the new single-particle density operator $\tilde{\rho}_T$, whose time development is given by Eq. (2.28). The solution of interest is that for $t=0$ when the electric field has reached its full value. Hence a solution of Eq. (2.28) must be found with the electric field given by Eq. (3.1) which reduces at $t=-\infty$ to the new equilibrium density operator, say $\tilde{\rho}$. This must satisfy Eq. (2.28) with H_E set equal to zero:

$$i\frac{\partial\tilde{\rho}}{\partial t}=[H_0+H'(\mathbf{r}),\tilde{\rho}]-\frac{e}{2mc}\times\sum_\alpha\{(\mathbf{p}\times\mathbf{B})_\alpha[x_\alpha,\tilde{\rho}]+[x_\alpha,\tilde{\rho}](\mathbf{p}\times\mathbf{B})_\alpha\}. \tag{3.2}$$

The calculation of the new density matrix will be limited to terms no higher than first order in the electric field. Therefore let

$$\tilde{\rho}_T=\tilde{\rho}_T+\tilde{\rho}_E, \tag{3.3}$$

where $\tilde{\rho}_E$ is taken to be linear in the electric field. Then the total density operator becomes equal to the equilibrium density operator when the electric field is zero. Inserting $\tilde{\rho}_T$ given by Eq. (3.3) into Eq. (2.28) and using Eq. (3.2), it is found that $\tilde{\rho}_E$ satisfies the equation

$$i\frac{\partial\tilde{\rho}_E}{\partial t}=[H_0+H'(\mathbf{r}),\tilde{\rho}_E]+[H_E,\tilde{\rho}]-\frac{e}{2mc}\sum_\alpha\{(\mathbf{p}\times\mathbf{B})_\alpha[x_\alpha,\tilde{\rho}_E]+[x_\alpha,\tilde{\rho}_E](\mathbf{p}\times\mathbf{B})_\alpha\} \tag{3.4}$$

if terms of second order in the electric field are neglected. The quantity $\tilde{\rho}_E$ must satisfy the initial condition

$$\tilde{\rho}_E(t=-\infty)=0. \tag{3.5}$$

Now since H_E may be written

$$H_E(\mathbf{r})=-eE_\alpha{}^0x_\alpha e^{st}, \tag{3.6}$$

Eqs. (3.4) and (3.5) can be satisfied by taking

$$\tilde{\rho}_E=fe^{st}, \tag{3.7}$$

where f is linear in the electric field and independent of the time. The quantity f is the correction to the equilibrium density operator at time zero, which is what is required. Inserting Eqs. (3.6) and (3.7) into Eq. (3.4) gives the equation for f,

$$isf=[H_0+H'(\mathbf{r}),f]-C-\frac{e}{2mc}\times\sum_\alpha\{(\mathbf{p}\times\mathbf{B})_\alpha[x_\alpha,f]+[x_\alpha,f](\mathbf{p}\times\mathbf{B})_\alpha\}, \tag{3.8}$$

where C is defined as the commutator

$$C=-eE_\alpha{}^0[\tilde{\rho},x_\alpha]. \tag{3.9}$$

To solve Eq. (3.8), which is valid in any representation, it is convenient to choose the representation for which H_0 is diagonal, that is, plane waves with periodic boundary conditions. The normalized eigenfunctions of H_0 are

$$\psi_k=(1/\sqrt{\Omega})e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{3.10}$$

where $\Omega=L^3$ is the volume of the container. The allowed

wave vectors are given by

$$k_\alpha = (2\pi/L)n_\alpha, \tag{3.11}$$

where n_α are all real positive and negative integers. Then ψ_k satisfies the equation

$$H_0\psi_k = \epsilon_k{}^0\psi_k,$$

where

$$\epsilon_k{}^0 = (1/2m)\mathbf{k}^2 \tag{3.12}$$

are the energy eigenvalues. In this representation Eq. (3.8) becomes

$$(\omega_{kk'}{}^0 - is)f_{kk'} = \sum_{k''}\{f_{kk''}H_{k''k'}' - H_{kk''}'f_{k''k'}\} + C_{kk'} + \frac{ie}{2mc}(\mathbf{k}+\mathbf{k}')\times\mathbf{B}\cdot(\nabla_k+\nabla_{k'})f_{kk'} \tag{3.13}$$

with $\omega_{kk'}{}^0 = \epsilon_k{}^0 - \epsilon_{k'}{}^0$. The matrix elements of $H'(\mathbf{r})$ are equal to

$$H_{kk'}' = \frac{1}{\Omega}\sum_{i=1}^{N} e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_i}\int_\Omega e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}}\phi(\mathbf{r})d\mathbf{r} \tag{3.14}$$

after making an appropriate change of variable. For simplicity the potential $\phi(\mathbf{r})$ will be taken to have a finite range so that the coordinate integration can be extended over all space. Then Eq. (3.14) becomes

$$H_{kk'}' = \frac{\phi_{kk'}}{\Omega}\sum_{i=1}^{N} e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_i}, \tag{3.15}$$

where

$$\phi_{kk'} = \int_{-\infty}^{\infty} e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}}\phi(\mathbf{r})d\mathbf{r}. \tag{3.16}$$

Following Kohn and Luttinger we separate Eq. (3.13) into diagonal and nondiagonal parts. The Eq. (3.13) becomes

$$-isf_k = \sum_{k'}'\{f_{kk'}H_{kk'}' - H_{kk'}'f_{k'k}\} + C_k + \frac{ie}{mc}(\mathbf{k}\times\mathbf{B})\cdot\nabla_k f_k \tag{3.17}$$

for $\mathbf{k}=\mathbf{k}'$ with $f_k \equiv f_{kk}$, $C_k \equiv C_{kk}$ and

$$(\omega_{kk'}-is)f_{kk'} = (f_k - f_{k'})H_{kk'}' + C_{kk'} + \sum_{k''}'\{f_{kk''}H_{k''k'}' - H_{kk''}'f_{k''k'}\} + \frac{ie}{2mc}(\mathbf{k}+\mathbf{k}')\times\mathbf{B}\cdot(\nabla_k+\nabla_{k'})f_{kk'} \tag{3.18}$$

for $\mathbf{k}\neq\mathbf{k}'$. Here terms in the sum having factors with pairs of equal indices have been separated out and advantage has been taken of the fact that H_{kk}' is a constant to define a new energy

$$\epsilon_k = \epsilon_k{}^0 + H_{kk}'$$

with

$$\omega_{kk'} = \epsilon_k - \epsilon_{k'}.$$

By resetting the zero of energy this constant can be removed. This is equivalent to taking $H_{kk}'=0$ and for simplicity it will be imagined that this has been done.

So far everything is quite general. It will now be shown that the technique of Kohn and Luttinger can be used to find a solution of Eq. (3.18) in a power series in λ. In developing such a solution it will be assumed that $\omega_0\tau_r$ is of order unity. This turns out to be a necessary condition for obtaining the ordinary Boltzmann transport equation when Eq. (3.17) is treated to lowest order in λ while retaining the magnetic field term. Since the collision relaxation time is inversely proportional to the square of the potential interaction of the electrons with the scattering centers, τ_r is of order λ^{-2} according to (2.3). For this reason factors of the magnetic field can conveniently be considered of order λ^2 in determining the respective orders of the various terms contained in Eq. (3.18).

To carry out the proposed development of the density matrix in powers of λ, the various terms in Eq. (3.18) must first be examined to learn how they depend on λ for small λ. First consider the commutator

$$C_{kk'} = -iE_\alpha{}^0[\tilde{\rho},x_\alpha]_{kk'} = ieE_\alpha{}^0\left(\frac{\partial}{\partial k_\alpha}+\frac{\partial}{\partial k_\alpha'}\right)\tilde{\rho}_{kk'}. \tag{3.19}$$

$\tilde{\rho}$ depends on the scattering potential and the magnetic field; $C_{kk'}$ can therefore be expanded in a power series in λ,

$$C_{kk'} = C_{kk'}{}^{(0)} + C_{kk'}{}^{(1)} + \cdots, \tag{3.20}$$

in which $C_{kk'}{}^{(n)}$ is of order λ^n. Here the evaluation of the series will be limited to the first term, $C_{kk'}{}^{(0)}$, since this turns out to be all that is required to obtain the lowest order result from Eq. (3.18).

In order to obtain the expansion indicated by Eq. (3.20) in powers of λ the corresponding expansion of $\tilde{\rho}$ is needed. $\tilde{\rho}$ satisfies the operator equation (3.2) from which the equation satisfied by $\tilde{\rho}^{(0)}$, the lowest order part of the new equilibrium density operator, can be obtained by setting $H'(\mathbf{r})$ and $\mathbf{B}$ equal to zero since they are of order λ and λ^2, respectively. Then

$$i\partial\tilde{\rho}^{(0)}/\partial t = [H_0,\tilde{\rho}^{(0)}]. \tag{3.21}$$

This is just the equation satisfied by the exact density operator ρ in the absence of both a magnetic field and scattering. In fact $\tilde{\rho}$ and ρ become identical in the absence of a magnetic field. Hence the solution of Eq. (3.21) at equilibrium is the usual Maxwell-Boltzmann function,

$$\rho^{(0)} = \tilde{\rho}^{(0)} = K^{(0)}e^{-\beta H_0}, \tag{3.22}$$

where

$$[K^{(0)}]^{-1} = \mathrm{Tr}e^{-\beta H_0}. \tag{3.23}$$

The matrix of (3.22) is just

$$\tilde{\rho}_{kk'}{}^{(0)} = \rho_k{}^0\delta_{kk'} \tag{3.24}$$

with

$$\rho_k^0=\frac{1}{\Omega}\left(\frac{2\pi\beta}{m}\right)^{3/2}e^{-\beta\epsilon_k}. \tag{3.25}$$

The first term in Eq. (3.20) is obtained by inserting (3.24) into (3.19). This gives

$$C_{kk'}{}^{(0)}=ieE_\alpha{}^0\frac{\partial\rho_k{}^0}{\partial k_\alpha}\delta_{kk'}. \tag{3.26}$$

Thus the diagonal terms of the commutator C begin with the zeroth order in λ while the nondiagonal ones are at least of first order in λ.

Now assume that f_k begins with order λ^{-2} as Kohn and Luttinger found when no magnetic field was present. This will lead to a consistent solution in the presence of a magnetic field as well. Then it follows from Eq. (3.18) and (3.26), on examination of the various terms, that $f_{kk'}(k\neq k')$ must begin with order λ^{-1}. Thus Eq. (3.18) can be solved by an iterative process. To lowest order Eq. (3.18) gives just

$$f_{kk'}{}^{(-1)}=[(f_k{}^{(-2)}-f_{k'}{}^{(-2)})/(\omega_{kk'}-is)]H_{kk'}, \tag{3.27}$$

in which appropriate superscripts have been added to indicate the respective orders in λ. Insert (3.27) along with (3.26) into Eq. (3.17) and again retain only lowest order in λ. This gives

$$\begin{aligned}-isf_k{}^{(-2)}=ieE_\alpha{}^0\frac{\partial\rho_k{}^0}{\partial k_\alpha}+\frac{ie}{mc}(\mathbf{k}\times\mathbf{B})\cdot\nabla_k f_k{}^{(-2)}\\+\sum_{k'}{}'|H_{kk'}|^2(f_k{}^{(-2)}-f_{k'}{}^{(-2)})\\\times\left\{\frac{1}{\omega_{kk'}-is}-\frac{1}{\omega_{kk'}+is}\right\}.\end{aligned} \tag{3.28}$$

From Eq. (3.15) it is found

$$|H_{kk'}|^2=\frac{|\phi_{kk'}|^2}{\Omega^2}\sum_{i=1}^{N}\sum_{j=1}^{N}e^{-i(\mathbf{r}_i-\mathbf{r}_j)\cdot(\mathbf{k}-\mathbf{k}')}. \tag{3.29}$$

This quantity depends in general on the positions of all the scatterers. Kohn and Luttinger showed, however, that the summation over k' effectively eliminates this dependence. If there is no correlation between the positions of the scatterers Eq. (3.28) becomes a well-defined equation independent of these positions. Kohn and Luttinger define the ensemble average of $M(\mathbf{r}_1,\mathbf{r}_2,\cdots\mathbf{r}_n)$, say $\langle M\rangle$, as the average of M over all different arrangements of the scatterers without any correlation between them. Then the ensemble average is given by the integral

$$\langle M\rangle=\frac{1}{\Omega^n}\int_\Omega\cdots\int_\Omega d\mathbf{r}_1\cdots d\mathbf{r}_n M(\mathbf{r}_1\cdots\mathbf{r}_n).$$

It can be shown without any loss of rigor that it is possible to replace $|H_{kk'}|^2$ in a sum such as occurs in Eq. (3.28) by its ensemble average given by

$$\langle|H_{kk'}|^2\rangle=\frac{N}{\Omega}|\phi_{kk'}|^2. \tag{3.30}$$

The solution of Eq. (3.28) still depends on the value of the frequency parameter s. There is, however, a large and useful range of s for which the solution is practically independent of s. This range is specified by the following conditions. First, s^{-1} must be much greater than the collision relaxation time, the cyclotron period, and the characteristic atomic time. When these conditions are satisfied the left-hand side of Eq. (3.28) may be dropped. Further if $s\gg\Delta E/\hbar$, where ΔE is of the order of the spacing of the translational electronic levels, the sum in Eq. (3.28) can be replaced by an integral according to

$$\sum_k=[\Omega/(2\pi)^3]\int d\mathbf{k}. \tag{3.31}$$

Then the condition that s be much less than characteristic atomic frequencies enables one to use the well-known result

$$\lim_{s\to+0}(x-is)^{-1}=P(1/x)+i\pi\delta(x), \tag{3.32}$$

where $P(1/x)$ is the principle value of $1/x$ and $\delta(x)$ is the Dirac delta function. Equation (3.31) can then be written

$$\begin{aligned}0=eE_\alpha{}^0\frac{\partial\rho_k{}^0}{\partial k_\alpha}+\frac{e}{mc}(\mathbf{k}\times\mathbf{B})\cdot\nabla_k f_k{}^{(-2)}\\+\frac{\Omega}{(2\pi)^3}\int\{W_{k'k}{}^{(2)}f_k{}^{(-2)}-W_{kk'}{}^{(2)}f_{k'}{}^{(-2)}\}d\mathbf{k}',\end{aligned} \tag{3.33}$$

where

$$w_{kk'}{}^{(2)}=(2\pi/\Omega^2)|\phi_{kk'}|^2\delta(\omega_{kk'}) \tag{3.34}$$

is the transition probability per unit time from a plane-wave state k to k' due to a single scattering event and $W_{kk'}{}^{(2)}=Nw_{kk'}{}^{(2)}$. Equation (3.33) is just the ordinary Boltzmann transport equation in the presence of a magnetic field. Notice that it is of over-all order λ^0. Once it has been solved for the diagonal part of the density matrix the nondiagonal elements can be obtained at once from Eq. (3.27). To calculate the average velocity the free-particle velocity operator $\mathbf{p}/m$ must be used. Here to lowest order the nondiagonal elements of the density matrix play no part since the velocity operator has only diagonal elements in a plane-wave representation.

IV. HIGHER APPROXIMATIONS

The problem of finding the higher order corrections to the Boltzmann transport equation (3.33) is considered in this section. To obtain such corrections it is only necessary to continue the iterative process begun in the last section. The process is carried only two orders further in λ since this is sufficient to bring in the first magnetic correction terms. With the exception of these terms the corrections are formally the same as those obtained by Kohn and Luttinger to this order. While there is in the present development an added implicit dependence of the density matrix on the magnetic field, the formal reduction of the corresponding terms, those not explicitly dependent on the magnetic field, remains the same and will not be repeated. Nonetheless some of the results of Kohn and Luttinger are necessary in order to properly understand the present work. In such cases the needed results will be quoted and referred to as K-L[3] with the corresponding equation number.

The diagonal elements of the density matrix f_k were found to begin with order λ^{-2} and will be needed to order λ^0. The nondiagonal elements $f_{kk'}$ begin with order λ^{-1} and will be needed to order λ. To the corresponding order Eqs. (3.17) and (3.18) become for very small s,

$$0=C_k^{(0)}+C_k^{(2)}+\frac{ie}{mc}(\mathbf{k}\times\mathbf{B})\cdot\nabla_k f_k+\sum_{k'}{}'(f_{kk'}H_{k'k}-H_{kk'}f_{k'k}), \quad (4.1)$$

$$(\omega_{kk'}-is)f_{kk'}=C_{kk'}^{(1)}+(f_k-f_{k'})H_{kk'}+\sum_{k''}{}'(f_{kk''}H_{k''k'}-H_{kk''}f_{k''k'})+\frac{ie}{2mc}(\mathbf{k}+\mathbf{k}')\times\mathbf{B}\cdot(\nabla_k+\nabla_{k'})f_{kk'}, \quad (4.2)$$

after inserting the commutator expansion (3.20) to the proper order. $C_k^{(1)}$ does not appear in Eq. (4.1) since it vanishes with the choice $H_{kk'}=0$. $C^{(0)}$ has no off-diagonal matrix elements according to Eq. (3.26) so that $C_{kk'}^{(0)}$ has not been included in Eq. (4.2).

It was shown in Sec. III that the lowest order solution of Eq. (4.2) is given by Eq. (3.27). If this is inserted in the last two terms of Eq. (4.2) and the resulting equation solved for $f_{kk'}$, the first correction is obtained. This process can be continued to obtain $f_{kk'}(k\neq k')$ as a power series in λ to any order. To the order already specified, the diagonal and nondiagonal matrix elements of the corrections to the equilibrium density matrix can be written

$$f_k=f_k^{(-2)}+f_k^{(-1)}+f_k^{(0)} \quad (4.3)$$

and

$$f_{kk'}=f_{kk'}^{(-1)}+f_{kk'}^{(0)}+f_{kk'}^{(1)}, \quad (4.4)$$

respectively. Here again the superscripts indicate the respective orders in λ. Placing Eqs. (4.3) and (4.4) in Eq. (4.2) it is readily found that $f_{kk'}^{(-1)}$ is given by Eq. (3.27) and

$$f_{kk'}^{(0)}=\frac{1}{\omega_{kk'}-is}\left[(f_k^{(-1)}-f_{k'}^{(-1)})H_{kk'}+\sum_{k''}{}'(kk''k')\left\{\frac{f_k^{(-2)}-f_{k''}^{(-2)}}{\omega_{kk''}-is}-\frac{f_{k''}^{(-2)}-f_{k'}^{(-2)}}{\omega_{k''k'}-is}\right\}\right], \quad (4.5)$$

$$f_{kk'}^{(1)}=\frac{1}{\omega_{kk'}-is}\left\{C_{kk'}^{(1)}+(f_k^{(0)}-f_{k'}^{(0)})H_{kk'}+\sum_{k''}{}'(kk''k')\left(\frac{f_k^{(-1)}-f_{k''}^{(-1)}}{\omega_{kk''}-is}-\frac{f_{k''}^{(-1)}-f_{k'}^{(-1)}}{\omega_{k''k'}-is}\right)\right.$$
$$+\sum_{\substack{k'',k'''\\ k''\neq k,k'\\ k'''\neq k,k''}}\frac{(kk'''k''k')}{\omega_{kk''}-is}\left(\frac{f_k^{(-2)}-f_{k'''}^{(-2)}}{\omega_{kk'''}-is}-\frac{f_{k'''}^{(-2)}-f_{k''}^{(-2)}}{\omega_{k'''k''}-is}\right)$$
$$-\sum_{\substack{k'',k'''\\ k''\neq k,k'\\ k\neq k'',k'}}\frac{(kk''k'''k')}{\omega_{k''k'}-is}\left(\frac{f_{k''}^{(-2)}-f_{k'''}^{(-2)}}{\omega_{k''k'''}-is}-\frac{f_{k'''}^{(-2)}-f_{k'}^{(-2)}}{\omega_{k'''k'}-is}\right)$$
$$\left.+\frac{ie}{2mc}(\mathbf{k}+\mathbf{k}')\times\mathbf{B}\cdot(\nabla_k+\nabla_{k'})\left(\frac{f_k^{(-2)}-f_{k'}^{(-2)}}{\omega_{kk'}-is}H_{kk'}\right)\right\}, \quad (4.6)$$

where

$$(kk''k')\equiv H_{kk''}H_{k''k'}$$
$$(kk'''k''k')\equiv H_{kk'''}H_{k'''k''}H_{k''k'}. \quad (4.7)$$

The last term in Eq. (4.6) is the most interesting; it is the first correction to the density matrix depending explicitly on the magnetic field. Except for the implicit dependence on the magnetic field, the remaining terms in Eqs. (4.5) and (4.6) are essentially the same as those obtained by Kohn and Luttinger.

Before finding the corresponding transport equation, it is convenient to rearrange the terms contained in $f_{kk'}(k\neq k')$ so that those of similar form can be combined. Add Eqs. (3.27), (4.5), and (4.6) and make use of Eq.

(4.3) to recombine the diagonal elements. Then $f_{kk'}(k\neq k')$ can be written

$$f_{kk'}=F_{kk'}{}^{(0)}+F_{kk'}{}^{(1)}+F_{kk'}{}^{(2)}, \tag{4.8}$$

where the F's are given by

$$F_{kk'}{}^{(0)}=\frac{(f_k-f_{k'})}{\omega_{kk'}-is}H_{kk'}, \tag{4.9}$$

$$F_{kk'}{}^{(1)}=\frac{1}{\omega_{kk'}-is}\sum_{k''}(kk''k')\left(\frac{f_k-f_{k''}}{\omega_{kk''}-is}-\frac{f_{k''}-f_{k'}}{\omega_{k''k'}-is}\right), \tag{4.10}$$

$$F_{kk'}{}^{(2)}=\frac{1}{\omega_{kk'}-is}\Bigg\{C_{kk'}{}^{(1)}+\sum_{\substack{k'',k'''\\ k''\neq k,k'\\ k'''\neq k,k''}}\frac{(kk'''k''k')}{\omega_{kk''}-is}\left(\frac{f_k-f_{k'''}}{\omega_{kk'''}-is}-\frac{f_{k'''}-f_{k''}}{\omega_{k'''k''}-is}\right)-\sum_{\substack{k'',k'''\\ k''\neq k,k'\\ k'''\neq k'',k'}}\frac{(kk''k'''k')}{\omega_{k''k'}-is}$$

$$\times\left(\frac{f_{k''}-f_{k'''}}{\omega_{k''k'''}-is}-\frac{f_{k'''}-f_{k'}}{\omega_{k'''k'}-is}\right)+\frac{ie}{2mc}(\mathbf{k}+\mathbf{k}')\times\mathbf{B}\cdot(\nabla_k+\nabla_{k'})\left(\frac{f_k-f_{k'}}{\omega_{kk'}-is}H_{kk'}\right)\Bigg\}. \tag{4.11}$$

$f_{kk'}(k\neq k')$, as expressed by the sum in Eq. (4.8) with f_k given by Eq. (4.3), differs from that of Eq. (4.4) only in terms of order λ^2 or higher, which are not being considered. It is apparent that $F_{kk'}$, defined by Eqs. (4.9) through (4.11), are actually mixtures of various orders in λ and correspond to Eqs. (K-L.34), (K-L.65), and (K-L.66).

To obtain the corrected transport equation insert Eq. (4.8) into Eq. (4.1); this gives

$$T_k{}^{(0)}+T_k{}^{(1)}+T_k{}^{(2)}=0, \tag{4.12}$$

where $T_k{}^{(0)}$ is just the right-hand side of Eq. (3.28) with $f_k{}^{(-2)}$ replaced by f_k. The two remaining terms in Eq. (4.12) are given by

$$T_k{}^{(1)}=\sum_{k'}{}'(F_{kk'}{}^{(1)}H_{k'k}'-H_{kk'}'F_{k'k}{}^{(1)}), \tag{4.13}$$

$$T_k{}^{(2)}=\sum_{k'}{}'(F_{kk'}{}^{(2)}H_{k'k}'-H_{kk'}'F_{k'k}{}^{(2)})+C_k{}^{(2)}. \tag{4.14}$$

If Eqs. (4.4) instead of Eq. (4.8) had been inserted in Eq. (4.1) the result would still have been Eq. (4.12) to terms of order λ^2. The introduction of the F's simply makes it possible to combine similar terms requiring complicated reductions. It should be clearly understood that the superscripts on $T_k{}^{(n)}$ and $F_{kk'}{}^{(n)}$ do not indicate their respective orders in λ as they do on all other symbols contained in this paper.[7]

Consider first Eq. (4.13) and replace $F_{kk'}{}^{(1)}$ using Eq. (4.10). This part of the transport equation is formally the same as Eq. (K-L.69) and can immediately be written

$$T_k{}^{(1)}=iN\sum_{k'}{}'(w_{k'k}{}^{(3)}f_k-w_{kk'}{}^{(3)}f_{k'}) \tag{4.15}$$

with $w_{kk'}{}^{(3)}$ given by Eq. (K-L.78).

We turn now to the consideration of Eq. (4.14). Three terms result: those linear in f_k, say $T_k{}^{(2)}(f)$, those containing the commutator, say $T_k{}^{(2)}(C)$, and those containing the magnetic field explicitly, say $T_k{}^{(2)}(\mathbf{B})$. Then Eq. (4.14) becomes

$$T_k{}^{(2)}=T_k{}^{(2)}(f)+T_k{}^{(2)}(C)+T_k{}^{(2)}(\mathbf{B}). \tag{4.16}$$

$T_k{}^{(2)}(f)$ is given by Eqs. (K-L.82), (K-L.83) and (K-L.84) and can therefore be written in the form

$$T_k{}^{(2)}(f)=iN\sum_{k'}{}'\{(w_{k'k}{}^{(4)}+u_{k'k}{}^{(4)}+v_{k'k}{}^{(4)})f_k$$

$$-(w_{kk'}{}^{(4)}+u_{kk'}{}^{(4)}+v_{kk'}{}^{(4)})f_{k'}\} \tag{4.17}$$

with $w_{kk'}{}^{(4)}$, $u_{kk'}{}^{(4)}$, and $v_{kk'}{}^{(4)}$, defined in terms of Eqs. (K-L.95), (K-L.96), and (K-L.98), respectively. The second term in Eq. (4.16) is equal to

$$T_k{}^{(2)}(C)=C_k{}^{(2)}+\sum_{k'}{}'\left\{\frac{C_{kk'}{}^{(1)}H_{k'k}'}{\omega_{kk'}-is}+\frac{H_{k'k}'C_{k'k}{}^{(1)}}{\omega_{kk'}+is}\right\} \tag{4.18}$$

which is formally the same as (K-L.81) except that C_k may depend explicitly on the magnetic field. The last term in the transport equation (4.16) takes the following form

$$T_k{}^{(2)}(\mathbf{B})=\frac{ie}{2mc}\sum_{k'}{}'|H_{kk'}|^2$$

$$\times\left\{\frac{1}{\omega_{kk'}-is}(\mathbf{k}+\mathbf{k}')\times\mathbf{B}\cdot(\nabla_k+\nabla_{k'})\frac{f_k-f_{k'}}{\omega_{kk'}-is}\right.$$

$$\left.-\frac{1}{\omega_{kk'}-is}(\mathbf{k}+\mathbf{k}')\times\mathbf{B}\cdot(\nabla_{k'}+\nabla_k)\frac{f_{k'}-f_k}{\omega_{kk'}-is}\right\}. \tag{4.19}$$

This term is new and depends explicitly on the magnetic field.

[7] The superscripts on the symbols contained in the paper of Kohn and Luttinger, Ref. 3, do not specify their true order in λ.

In order to calculate $T_k^{(2)}(C)$ the transformed equilibrium density matrix, $\tilde{\rho}_{kk'}$, must first be found to second order in λ. The density operator $\tilde{\rho}$ itself can be found in a quite straightforward way by placing the exact, known, equilibrium density operator (2.8) in Eq. (2.11):

$$K\exp\{-\beta[H(\mathbf{p}-(e/c)\mathbf{A})+H'(\mathbf{r})]\}$$
$$=\int e^{i\mathbf{P}\cdot\xi}R(\xi,\mathbf{r},\mathbf{B})d\xi. \quad (4.20)$$

Once R is found from this relationship it can be inserted in Eq. (2.26) to obtain $\tilde{\rho}$ directly. However, since the zero-order solution is now known (see Sec. III), it is much easier to obtain $\tilde{\rho}_{kk'}$ by a process of iteration from the matrix equation satisfied by it:

$$\omega_{kk'}\tilde{\rho}_{kk'}=(\tilde{\rho}_k-\tilde{\rho}_{k'})H_{kk'}'$$
$$+\sum_{k''(\neq k,k')}\{\tilde{\rho}_{kk''}H_{k''k'}'-H_{kk''}'\tilde{\rho}_{k''k'}\}$$
$$+\frac{ie}{2mc}(\mathbf{k}+\mathbf{k}')\times\mathbf{B}\cdot(\nabla_k+\nabla_{k'})\tilde{\rho}_{kk'}. \quad (4.21)$$

This equation follows directly from Eq. (3.2) since at equilibrium the time derivative of $\tilde{\rho}$ must vanish. The normalization of $\tilde{\rho}$, which can be found from Eq. (2.39), depends in general on the scattering potential and the magnetic field so that care must be taken to correctly include both types of terms. Notice, however, that the differential operator in Eq. (4.21) gives nothing when it acts on the zero-order solution Eq. (3.24). Therefore, to terms of order λ^2, no corrections to $\tilde{\rho}_{kk'}$ exist which are linear in the magnetic field. Terms containing the magnetic field can be completely disregarded in determining $\tilde{\rho}_{kk'}$ at least to order λ^2. By carrying out the iterative process using Eq. (3.24) as the zero-order solution it can readily be shown that the result is identical with that obtained by Kohn and Luttinger using a quite different procedure, namely, that implied by Eqs. (K-L.C6) and (K-L.C7). Hence to order λ^2, C_k is still given by (K-L.C6) even in the presence of a magnetic field. This implies at once that $T_k^{(2)}(C)$ remains unchanged in the presence of a magnetic field and is still given by Eq. (K-L.87).

Thus the corrections to the Boltzmann equation depending implicitly on the magnetic field are formally the same as those found by Kohn and Luttinger in the absence of a magnetic field and it is possible to take over their results practically intact. However, the term $T_k^2(\mathbf{B})$ given by Eq. (4.19) is completely new and depends explicitly on the magnetic field. If the sums contained in Eq. (4.19) are replaced by integrals using the transformation (3.31) and $|H_{kk'}'|^2$ is replaced by its ensemble average, it can be rewritten

$$T_k^{(2)}(\mathbf{B})=\frac{ien}{c(2\pi)^3}\Big\{\int|\phi_{kk'}|^2(\nabla_k f_k-\nabla_{k'}f_{k'})$$
$$\cdot\mathbf{B}\times(\nabla_k-\nabla_{k'})\frac{1}{\omega_{kk'}}d\mathbf{k}'+\int|\phi_{kk'}|^2(f_k-f_{k'})$$
$$\times\mathbf{B}\cdot(\nabla_k\times\nabla_{k'})\frac{1}{\omega_{kk'}}d\mathbf{k}'\Big\}, \quad (4.22)$$

after some rearrangement of the derivative operators. Here the density of scattering centers has been set equal to n. The integrals contained in Eq. (4.22) are meaningless as they stand since derivative operators still act on the reciprocal of $\omega_{kk'}$ giving divergent integrals. If f_k and its first derivatives vanish at infinity, then partial integrations can be performed which allow Eq. (4.22) to be rewritten as a set of principal value integrals

$$T_k^{(2)}(\mathbf{B})=\frac{ien}{c(2\pi)^3}\Big\{\nabla_k f_k\cdot\mathbf{B}\times\nabla_k\int P\frac{|\phi_{kk'}|^2}{\omega_{kk'}}d\mathbf{k}'$$
$$+\mathbf{B}\times\nabla_k\cdot\int P\frac{(\nabla_k|\phi_{kk'}|^2)}{\omega_{kk'}}(f_k-f_{k'})d\mathbf{k}'$$
$$\times\int P\frac{(\nabla_k f_k-\nabla_{k'}f_{k'})}{\omega_{kk'}}\cdot\mathbf{B}\times\nabla_k|\phi_{kk'}|^2d\mathbf{k}'\Big\}. \quad (4.23)$$

$T_k^{(2)}(\mathbf{B})$ is a new set of terms in the transport equation arising from the interference between the change in the distribution function caused by the presence of the magnetic field and the change brought about by collisions of electrons with scattering centers. The contribution of these terms to the current density will be considered in a later publication.

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APPENDIX

Let $\mathbf{P}=(\mathbf{p}-(e/c)\mathbf{A}(\mathbf{r}))$, where $\mathbf{p}$ is the ordinary momentum operator and $\mathbf{A}(\mathbf{r})$ is the vector potential. Then the Fourier integral

$$\int e^{i\mathbf{P}\cdot\xi}F(\xi,\mathbf{r})d\xi=0 \quad (A1)$$

implies that $F(\xi,\mathbf{r})$ must vanish. In order to show this choose for simplicity a Landau gauge, $\mathbf{A}(\mathbf{r}) = (-yB, 0, 0)$. Then (A1) can be written

$$\int e^{i\mathbf{p}\cdot\xi} e^{im\omega_0\xi_x y} e^{-\frac{1}{2}im\omega_0\xi_x\xi_y} F(\xi,\mathbf{r})d\xi = 0 \qquad \text{(A2)}$$

by using Eq. (2.18). Form the matrix of (A2) using a plane-wave representation (see Sec. III). Then (A2) becomes

$$\int e^{i\mathbf{k}\cdot\xi} F_{kk'}'(\xi,\mathbf{r})d\xi = 0, \qquad \text{(A3)}$$

where $F'(\xi,\mathbf{r})$ is defined by

$$F'(\xi,\mathbf{r}) = e^{im\omega_0\xi_x y} e^{-\frac{1}{2}im\omega_0\xi_x\xi_y} F(\xi,\mathbf{r}). \qquad \text{(A4)}$$

Equation (A3) is now an ordinary Fourier integral and implies that $F'(\xi,\mathbf{r})$ must be zero. According to (A4), this in turn requires that $F(\xi,\mathbf{r})$ must vanish.

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Nonlinear Susceptibility Constants and Self-Focusing of Optical Beams in Liquids*

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This paper reports measurements of intensity-induced rotation of the polarization ellipse, and measurements of the self-focusing threshold and its polarization dependence, using an unfocused laser beam in several Raman-active liquids. An attempt was made to determine the nonlinear susceptibility constants in these liquids by correlating these measurements. It is found that the self-focusing formulas for linearly polarized beams are in good agreement with the experiments, but the polarization dependence of the self-focusing threshold disagrees with the corresponding polarization dependence of the nonlinear index changes. A possible explanation for this discrepancy is discussed.

I. INTRODUCTION

IT is *assumed* in linear optics that a light beam of finite cross section can be represented by a superposition of unbounded plane-wave components propagating in slightly different directions.[1] The presence of optical nonlinearities produced by intense laser beams in a dielectric medium invalidates the principle of superposition. The plane-wave components are no longer independent, but are coupled to each other through the nonlinear polarization terms which bring about transfer of energy among the components.[2] The nonlinear optical effects of an intense laser beam on the propagation of the beam itself have been considered by several authors;[3–6] they include the intensity-induced rotation of the polarization ellipse,[5,6] and the intensity-induced slowing of the plane-wave components, which leads to self-focusing of the laser beam.[7,8]

This paper discusses some of the nonlinear optical effects which are related to the intensity-dependent changes in the real part of the index of refraction. In Sec. II, a phenomenological description is first given of the self-induced effects of an unbounded plane wave in a medium which is lossless and isotropic in the linear approximation. To describe these effects, a fourth-rank nonlinear susceptibility tensor with three nonzero independent components is introduced. Additional nonlinear effects associated with laser beams of finite cross section are then discussed in Sec. III. Possible ways to determine the susceptibility constants are discussed in Sec. IV and the relations that exist between these susceptibility constants are derived in Sec. V in terms of a simplified physical model.

Section VII reports measurements of intensity-induced rotation of the polarization ellipse associated with an unfocused laser beam in several Raman-active liquids. These measurements were correlated with measurements of self-focusing threshold[9] to determine the nonlinear susceptibility constants in these liquids.

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[1] See, for example, F. A. Jenkins and H. E. White, *Fundamentals of Optics* (McGraw-Hill Book Company, Inc., New York, 1957), Chap. 12.

[2] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).

[3] R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Letters **13**, 479 (1964).

[4] V. I. Talanov, JETP Pis'ma Redaktsiyu **2**, 218 (1965) [English transl.: Soviet Phys.—JETP Letters **2**, 138 (1965)].

[5] P. D. Maker, R. W. Terhune, and C. M. Savage, Phys. Rev. Letters **12**, 507 (1964).

[6] P. D. Maker and R W. Terhune, Phys. Rev. **137**, A801 (1965).

[7] M. Hercher, J. Opt. Soc. Am. **56**, 563 (1964).

[8] N. F. Pilipetskii and A. R. Rustamov, JETP Pis'ma Redaktsiyu **2**, 88 (1965) [English transl.: Soviet Phys.—JETP Letters **2**, 55 (1965)].

[9] P. L. Kelley, Phys. Rev. Letters **15**, 1005 (1965).