Conductivity Tensor of an Anisotropic Quantum Plasma in a Uniform Magnetic Field

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(Received 17 January 1964; revised manuscript received 10 March 1964)

The quantum magnetoconductivity tensor for a system in which the constant energy surfaces are ellipsoids tilted away from the direction of the dc magnetic field is derived. The results are applied to a system with cubic symmetry in which the Fermi surface consists of a set of ellipsoids. The dispersion relation for low-frequency magnetoplasma oscillations (helicons) is investigated, and some qualitative discussion of the attenuation of helicon waves in anisotropic materials is presented.

I. INTRODUCTION

THE degenerate electron gas model[1] has been extremely useful in the study of the electronic properties of metals, semimetals, and degenerate semiconductors in the presence of a uniform magnetic field. The qualitative understanding of the magnetic field dependence of the attenuation[2,3] and velocity[4] of sound waves, of the microwave surface impedance,[5] and of the propagation[6,7] of helicon and Alfvén waves in solids attests to the usefulness of the model. However, the constant energy surfaces in wave-vector space for real solids often differ considerably from the simple spherical surfaces of the degenerate electron gas. It is conceivable that with more complicated energy surfaces entirely new effects,[8] which are not present in the simple spherical energy surface model, can occur. In order to investigate the possibility of such new effects, we consider a system in which the constant energy surfaces are ellipsoids tilted at an arbitrary angle to the direction of the dc magnetic field. It should be pointed out that there exist a number of materials in which the constant energy surfaces are well approximated by a set of ellipsoids. Bismuth is a classic example of such a material.

In Sec. II of this paper we study the quantum magnetoconductivity tensor for a system in which the Fermi surface is a single ellipsoid. The principal axes of the ellipsoid are not necessarily oriented parallel or perpendicular to the direction of the dc magnetic field. If the Fermi surface consists of a set of ellipsoids, the conductivity tensor can be approximated by the sum of the conductivity tensors for each ellipsoid. In the final section we consider a multiellipsoid system which has cubic symmetry in the absence of the dc magnetic field. Some general conclusions concerning the propagation and attenuation of helicon waves are made by inspecting the form of the conductivity tensor.

II. CONDUCTIVITY TENSOR

In this section we consider a rather unrealistic model, in which the Fermi surface is a single ellipsoid. The Hamiltonian for an electron in the absence of the dc magnetic field is assumed to be

$$\mathcal{H}_{00}=\frac{1}{2m}\{\beta_1 p_{x'}^2+\beta_2 p_{y'}^2+\beta_3 p_{z'}^2\}. \tag{1}$$

Here we have chosen a Cartesian coordinate system (x',y',z') with each of the coordinate axes parallel to one of the principal axes of the ellipsoid. We now introduce a dc magnetic field of induction $\mathbf{B}_0$. For simplicity we choose $\mathbf{B}_0$ to lie in the y'-z' plane, and we define θ as the angle between the z' axis and $\mathbf{B}_0$. The completely general problem (where $\mathbf{B}_0$ is not restricted to be in the y'-z' plane) can also be solved, but no new physical effects arise from the generalization. The Hamiltonian for a single electron in the presence of the dc magnetic field can be written[9]

$$\mathcal{H}_0=\frac{1}{2m}\left\{\alpha_1 p_x^2+\alpha_2\left(p_y-\frac{e}{c}B_0x\right)^2+\alpha_3 p_z^2+2\alpha_4\left(p_y-\frac{e}{c}B_0x\right)p_z\right\}. \tag{2}$$

In this equation we have introduced a new Cartesian coordinate system (x,y,z) which is obtained by rotating the (x',y',z') system about the x' axis so that $\mathbf{B}_0$ is in the z direction. We have also taken $\mathbf{A}_0=(0,B_0x,0)$ for the vector potential of the dc magnetic field of induction $\mathbf{B}_0$. The parameters α_1 through α_4 are related to β_1

[1] See for example J. J. Quinn, Arkiv Fysik **26**, 93 (1964) for a discussion of the degenerate electron gas model and for a review of some of the applications of this model to properties of metals in the presence of a uniform magnetic field.
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[8] One such effect is the parallel field magnetoacoustic effect observed by L. Mackinnon, M. T. Taylor, and M. R. Daniels, Phil. Mag. **7**, 523 (1962). For a discussion of the origin of this effect see M. R. Daniels and L. Mackinnon, *ibid.* **8**, 537 (1963); and J. J. Quinn, Phys. Rev. Letters **11**, 316 (1963).
[9] See J. M. Luttinger and W. Kohn, Phys. Rev. **97**, 869 (1955).

through β_3 by the equations

$$\begin{aligned}\alpha_1&=\beta_1,\\ \alpha_2&=\beta_2\cos^2\theta+\beta_3\sin^2\theta,\\ \alpha_3&=\beta_2\sin^2\theta+\beta_3\cos^2\theta,\\ \alpha_4&=(\beta_2-\beta_3)\cos\theta\sin\theta.\end{aligned}\tag{3}$$

The stationary states of the Hamiltonian $\mathcal{H}_0$ are characterized by the eigenfunctions

$$|\nu\rangle=|nk_yk_z\rangle=L^{-1}\exp(ik_yy+ik_zz)\times u_n\left(x+\frac{\hbar[\alpha_1\alpha_2]^{1/2}}{m\omega_0}\left[k_y+\frac{\alpha_4}{\alpha_2}k_z\right]\right),$$

and the eigenvalues (4)

$$E_\nu=E_n(k_z)=\hbar\omega_0(n+\tfrac{1}{2})+(\alpha_3-\alpha_4^2/\alpha_2)\hbar^2k_z^2/2m.$$

In Eqs. (3) and (4) $\omega_0=(\alpha_1\alpha_2)^{1/2}|e|B_0/mc$ is the cyclotron frequency of the electrons, and $u_n(x)$ is a normalized simple harmonic oscillator wave function for a particle of mass $\alpha_1^{-1}m$ and characteristic frequency ω_0. The allowed values of the wave vectors k_y and k_z are determined by imposing periodic boundary conditions in the y and z directions (we have assumed the sample is contained in a cubic box of volume L^3). The quantum number n can take on any nonnegative integral value.

The introduction of an electromagnetic disturbance which varies as $\exp(i\omega t-i\mathbf{q}\cdot\mathbf{r})$ gives rise to a self-consistent electromagnetic field. The self-consistent field can be described in terms of a vector potential $\mathbf{A}(\mathbf{r},t)$ and a scalar potential $\phi(\mathbf{r},t)$. For simplicity we choose a gauge in which $\phi=0$; then to first order in $\mathbf{A}$ the Hamiltonian for an electron in the presence of the dc magnetic field and self-consistent field is

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1.\tag{5}$$

$\mathcal{H}_0$ is defined by Eq. (2) and $\mathcal{H}_1$ is given by

$$\mathcal{H}_1=-\frac{e}{2c}(\mathbf{v}\cdot\mathbf{A}+\mathbf{A}\cdot\mathbf{v}).\tag{6}$$

The operator $\mathbf{v}=i\hbar^{-1}[\mathcal{H}_0,\mathbf{r}]$ is the velocity operator for an electron in the field $\mathbf{B}_0$; the components of $\mathbf{v}$ are given by

$$\begin{aligned}v_x&=m^{-1}\alpha_1p_x,\\ v_y&=m^{-1}\alpha_2[p_y+(\alpha_1\alpha_2)^{-1/2}m\omega_0x]+m^{-1}\alpha_4p_z,\\ v_z&=m^{-1}\alpha_4[p_y+(\alpha_1\alpha_2)^{-1/2}m\omega_0x]+m^{-1}\alpha_3p_z.\end{aligned}\tag{7}$$

The electron current density induced by the self-consistent field is obtained by taking the trace of the product of the current density operator and the single particle density matrix. The density matrix ρ is set equal to $\rho_0+\rho_1$, where ρ_0 is the value of ρ in the absence of the perturbing self-consistent field, and ρ_1 is the small change in ρ from this equilibrium value cause by the self-consistent field. The operator ρ satisfies the equation of motion

$$i\hbar(\partial/\partial t)\rho=[\mathcal{H},\rho],\tag{8}$$

where $[\mathcal{H},\rho]$ means the commutator of $\mathcal{H}$ with ρ. If we assume ρ_1 varies as $\exp(i\omega t)$ and neglect terms of higher than first order in $\mathbf{A}$, Eq. (8) can be rewritten

$$-\hbar\omega\rho_1=[\mathcal{H}_0,\rho_1]+[\mathcal{H}_1,\rho_0].\tag{9}$$

By taking off-diagonal matrix elements of Eq. (9) in the representation defined by Eqs. (4) one obtains the equation

$$\langle\nu|\rho_1|\nu'\rangle=[f_0(E_{\nu'})-f_0(E_\nu)](E_{\nu'}-E_\nu-\hbar\omega)^{-1}\times\langle\nu|\mathcal{H}_1|\nu'\rangle.\tag{10}$$

In obtaining Eq. (10) we have made use of the relation $\rho_0|\nu\rangle=f_0(E_\nu)|\nu\rangle$, where $f_0(E_\nu)$ is the Fermi distribution function for the energy E_ν. The current density operator, $\mathbf{j}_{\mathrm{op}}=(ie/\hbar)[\mathcal{H},\mathbf{r}]$, can be written

$$\mathbf{j}_{\mathrm{op}}=e\mathbf{v}-(e^2/mc)\boldsymbol{\alpha}\cdot\mathbf{A},\tag{11}$$

where

$$\boldsymbol{\alpha}=\begin{bmatrix}\alpha_1&0&0\\0&\alpha_2&\alpha_4\\0&\alpha_4&\alpha_3\end{bmatrix}.\tag{12}$$

The current density induced by the self-consistent field at a point $\mathbf{r}_0$ and time t is

$$\mathbf{j}(\mathbf{r}_0,t)=\sum_\nu\langle\nu|\tfrac{1}{2}\{\mathbf{j}_{\mathrm{op}}\delta(\mathbf{r}-\mathbf{r}_0)+\delta(\mathbf{r}-\mathbf{r}_0)\mathbf{j}_{\mathrm{op}}\}\rho|\nu\rangle.\tag{13}$$

We introduce the operator $\mathbf{V}(\mathbf{q})$ defined by the equation

$$\mathbf{V}(\mathbf{q})=\tfrac{1}{2}(\mathbf{v}e^{i\mathbf{q}\cdot\mathbf{r}}+e^{i\mathbf{q}\cdot\mathbf{r}}\mathbf{v}).\tag{14}$$

The Fourier transform $\mathbf{j}(\mathbf{q},\omega)$ of the induced current density $\mathbf{j}(\mathbf{r}_0,t)$ turns out to be

$$\mathbf{j}(\mathbf{q},\omega)=\frac{e}{\Omega}\sum_{\nu\nu'}\langle\nu'|\mathbf{V}(\mathbf{q})|\nu\rangle\langle\nu|\rho_1|\nu'\rangle-\frac{e^2N}{mc\Omega}\boldsymbol{\alpha}\cdot\mathbf{A}(\mathbf{q},\omega).\tag{15}$$

In Eq. (15) $\mathbf{A}(\mathbf{q},\omega)$ is the Fourier transform of $\mathbf{A}(\mathbf{r},t)$ and Ω and N are the volume and the total number of electrons in the system. By using Eqs. (10) and (6), $\mathbf{j}(\mathbf{q},\omega)$ can be expressed as

$$\mathbf{j}(\mathbf{q},\omega)=-\frac{e^2N}{mc\Omega}\left\{\boldsymbol{\alpha}+\frac{m}{N}\sum_{\nu\nu'}\frac{f_0(E_{\nu'})-f_0(E_\nu)}{E_{\nu'}-E_\nu-\hbar\omega}\times\langle\nu'|\mathbf{V}(\mathbf{q})|\nu\rangle\langle\nu'|\mathbf{V}(\mathbf{q})|\nu\rangle^*\right\}\cdot\mathbf{A}(\mathbf{q},\omega).\tag{16}$$

The electric field strength $\mathbf{E}$ and the magnetic induction $\mathbf{B}$ of the self-consistent field are simply $(-i\omega/c)\mathbf{A}$ and $-i\mathbf{q}\times\mathbf{A}$. The conductivity tensor is defined by the relation

$$\mathbf{j}(\mathbf{q},\omega)=\boldsymbol{\sigma}(\mathbf{q},\omega)\cdot\mathbf{E}(\mathbf{q},\omega).\tag{17}$$

From Eq. (16) it can readily be seen that

$$\boldsymbol{\sigma}(\mathbf{q},\omega)=\frac{\omega_p^2}{4\pi i\omega}\left\{\boldsymbol{\alpha}+\frac{m}{N}\sum_{\nu\nu'}\frac{f_0(E_{\nu'})-f_0(E_\nu)}{E_{\nu'}-E_\nu-\hbar\omega}\times\langle\nu'|\mathbf{V}(\mathbf{q})|\nu\rangle\langle\nu'|\mathbf{V}(\mathbf{q})|\nu\rangle^*\right\}, \quad (18)$$

where $\omega_p^2=4\pi Ne^2/\Omega m$. In order to calculate the matrix elements of the operator $\mathbf{V}(\mathbf{q})$, it is convenient to write the components of $\mathbf{V}(\mathbf{q})$ as follows:

$$\begin{aligned} V_x(\mathbf{q})&=e^{i\mathbf{q}\cdot\mathbf{r}}[v_x+\hbar\alpha_1 q_x/2m],\\ V_y(\mathbf{q})&=e^{i\mathbf{q}\cdot\mathbf{r}}[v_y+\hbar(\alpha_2 q_y+\alpha_4 q_z)/2m],\\ V_z(\mathbf{q})&=e^{i\mathbf{q}\cdot\mathbf{r}}[v_z+\hbar(\alpha_4 q_y+\alpha_3 q_z)/2m]. \end{aligned} \quad (19)$$

In Eq. (19), v_x, v_y, and v_z are the components of the velocity operator defined by Eq. (7). It is straightforward but tedious to show that

$$\langle n'k_y'k_z'|\mathbf{V}(\mathbf{q})|nk_yk_z\rangle=\delta(k_y',k_y+q_y)\delta(k_z',k_z+q_z)\times\exp\left\{-iq_x\frac{\hbar(\alpha_1\alpha_2)^{1/2}}{m\omega_0}\left(k_y+\frac{\alpha_4}{\alpha_2}k_z\right)\right\}\mathbf{F}_{n'n}(\mathbf{q}). \quad (20)$$

In Eq. (20), $\delta(k',k)=0$ unless $k'=k$ when it has the value unity. The components of $\mathbf{F}_{n'n}(\mathbf{q})$ are given by

$$\begin{aligned} [F_{n'n}(\mathbf{q})]_x&=i\left(\frac{\alpha_1\hbar\omega_0}{2m}\right)^{1/2}X_{n'n}^{(-)}(\mathbf{q})+\frac{\hbar}{2m}\alpha_1 q_x f_{n'n}(\mathbf{q}),\\ [F_{n'n}(\mathbf{q})]_y&=\left(\frac{\alpha_2\hbar\omega_0}{2m}\right)^{1/2}X_{n'n}^{(+)}(\mathbf{q})+\frac{\hbar}{2m}(\alpha_2 q_y+\alpha_4 q_z)f_{n'n}(\mathbf{q}),\\ [F_{n'n}(\mathbf{q})]_z&=\left(\frac{\alpha_4^2}{\alpha_2}\frac{\hbar\omega_0}{2m}\right)^{1/2}X_{n'n}^{(+)}(\mathbf{q})+\frac{\hbar}{m}\left[\left(\alpha_3-\frac{\alpha_4^2}{\alpha_2}\right)\times k_z+\frac{\alpha_4}{2}q_y+\frac{\alpha_3}{2}q_z\right]f_{n'n}(\mathbf{q}). \end{aligned} \quad (21)$$

The symbol $f_{n'n}(\mathbf{q})$ is defined by the equation

$$f_{n'n}(\mathbf{q})=\int_{-\infty}^{\infty}dx u_n'\left(x+\frac{\hbar}{m\omega_0}[\alpha_1\alpha_2]^{1/2}\times\left[q_y+\frac{\alpha_4}{\alpha_2}q_z\right]\right)e^{iq_x x}u_n(x), \quad (22)$$

and the quantities $X_{n'n}^{(\pm)}(\mathbf{q})$ are related to the $f_{n'n}(\mathbf{q})$ by the equation

$$X_{n'n}^{(\pm)}(\mathbf{q})=(n+1)^{1/2}f_{n',n+1}(\mathbf{q})\pm n^{1/2}f_{n',n-1}(\mathbf{q}). \quad (23)$$

Some useful properties of the functions $f_{n'n}(\mathbf{q})$ and $X_{n'n}^{(\pm)}(\mathbf{q})$ are given in the Appendix. By using Eqs. (20), the expression for the conductivity tensor can be written

$$\boldsymbol{\sigma}(\mathbf{q},\omega)=\frac{\omega_p^2}{4\pi i\omega}\left\{\boldsymbol{\alpha}+\frac{m}{N}\times\sum_{n'nk_yk_z}\frac{f_0[E_{n'}(k_z+q_z)]-f_0[E_n(k_z)]}{E_{n'}(k_z+q_z)-E_n(k_z)-\hbar\omega}\times\mathbf{F}_{n'n}(\mathbf{q})\mathbf{F}_{n'n}^*(\mathbf{q})\right\}. \quad (24)$$

III. CONDUCTIVITY TENSOR FOR A SET OF ELLIPSOIDS

Thus far we have not included the scattering of electrons by thermal phonons or lattice imperfections. For a simple spherical Fermi surface this can be accomplished by introducing a phenomenological relaxation time τ. The only change in the conductivity[3] tensor caused by the introduction of collisions is the replacement of ω by $\omega-i/\tau$. For a system in which the Fermi surface consists of a number of ellipsoids, it is not obvious that the effect of collisions can be accounted for by simply replacing ω by $\omega-i/\tau$; however, in what follows, we shall assume that it can. Further, we shall assume that the self-consistent field does not affect the population of any of the ellipsoids. With these simplifications the conductivity for the system of ellipsoids is simply the sum of the conductivities of the individual ellipsoids.

$$\boldsymbol{\sigma}(\mathbf{q},\omega)=\sum_j\boldsymbol{\sigma}^{(j)}(\mathbf{q},\omega), \quad (25)$$

where $\boldsymbol{\sigma}^{(j)}$ is the conductivity tensor for the jth ellipsoid.

As a simple example to which the conductivity tensor discussed in this work is applicable, we consider helicon propagation in a many-ellipsoid system which has cubic symmetry in the absence of the dc magnetic field. If the magnetic field is in a symmetry direction, the only off-diagonal elements of $\boldsymbol{\sigma}$ are $\sigma_{xy}=-\sigma_{yx}$. For propagation parallel to the dc magnetic field, it is convenient to introduce the conductivities $\sigma_\pm$ defined by the relation

$$\sigma_\pm=\sigma_{xx}\mp i\sigma_{xy}. \quad (26)$$

$\sigma_\pm$ is the sum of contributions from each ellipsoid. Using Eq. (24), one can easily see that for the jth ellipsoid

$$\sigma_\pm^{(j)}(\mathbf{q},\omega)=\frac{\omega_p^2\alpha_1}{4\pi i\omega}\left\{1+\frac{\hbar\omega_0}{2N}\times\sum_{n'nk_yk_z}\left[\frac{f_0[E_{n'}(k_z+q)]-f_0[E_n(k_z)]}{E_{n'}(k_z+q)-E_n(k_z)-\hbar\omega}\times X_{n'n}^{(-)}\left(X_{n'n}^{(-)}\pm[\alpha_2/\alpha_1]^{1/2}X_{n'n}^{(+)}+\left[\frac{\alpha_4^2}{\alpha_1}\frac{\hbar q^2}{2m\omega_0}\right]^{1/2}f_{n'n}\right)\right]\right\}, \quad (27)$$

where all the α_i, ω_0, etc., are those values appropriate to the jth ellipsoid. In this equation ω must be replaced by $\omega - i/\tau$ to account for collisions. In the limit of infinite wavelength (i.e., $q \to 0$) $\sigma_\pm^{(j)}$ reduces to the expression

$$\sigma_\pm^{(j)} = -\frac{\omega_p^2}{4\pi i}\frac{\alpha_1(\omega-(i/\tau)\pm\alpha_2|e|B_0/mc)}{\alpha_1\alpha_2(|e|B_0/mc)^2-(\omega-i/\tau)^2}. \quad (28)$$

For $\alpha_k|e|B_0/mc \gg |\omega - i/\tau|$ for $k=1$ and 2, $\sigma_\pm^{(j)}$ is almost purely imaginary and depends only on the number of electrons in the ellipsoid and the strength of the dc magnetic field. Thus the frequency of low-frequency magnetoplasma oscillations,[10] which is given by

$$\omega^2 = c^2q^2\left[1-\frac{4\pi i}{\omega}\sigma_+\right]^{-1}, \quad (29)$$

will be independent of the direction of the magnetic field and of the effective masses of the carrier. In this limit the helicon frequency is $\omega_H = q^2cB_0/4\pi|e|n$, where n is the total electron density of the solid.

Because $\sigma_\pm(q,\omega)$ is a sum of terms like $\sigma_\pm^{(j)}$ given in Eq. (27), some interesting observations can be made simply by studying the form of $\sigma_\pm^{(j)}$. In the case of a spherical Fermi surface, the functions $X_{n'n}^{(\pm)}$ reduce to $(n+1)^{1/2}\delta_{n',n+1} \pm n^{1/2}\delta_{n',n-1}$ for propagation parallel to $\mathbf{B}_0$. Thus only terms with $n'=n\pm 1$ contribute to the summation. In the limit that $\omega_0\tau \gg 1$, this results in essentially no attenuation of helicon waves (real part of $\sigma_\pm$ is zero) until qv_F, the product of the wave vector of the helicon and the Fermi velocity, becomes equal to ω_0. This is just the well-known threshold for the Doppler-shifted cyclotron absorption.

For an ellipsoidal energy surface, however, $X_{n'n}^{(\pm)}$ reduces to a sum of Kronicker delta functions only if *all the off-diagonal components of the tensor* $\boldsymbol{\alpha}$ *vanish.* Thus for an arbitrarily oriented ellipsoid, one can have transitions in which $n'=n$. There is no threshold for these transitions; they are present even at the lowest frequencies. The existence of $\Delta n=0$ transitions might lead to an additional broadening of the dimensional resonances of helicons[11] in materials like lead telluride, whose Fermi surface consists of a set of ellipsoids. Furthermore the attenuation of the helicon waves due to these transitions will show "geometric resonances." The physical origin of these geometric resonances is exactly the same as that of the parallel field magnetoacoustic effect.[8] The projection of the cyclotron orbit of the electrons onto the direction of propagation can match an integral or half-integral number of helicon wavelengths. The mathematical origin of these "geometric resonances" can easily be seen by writing down the expression for the real part of $\sigma_\pm^{(j)}$.

$$\mathrm{Re}\sigma_\pm^{(j)} \propto \sum_{n\beta}\Big\{f_0[E_n(\kappa_\beta)+\hbar\omega]-f_0[E_n(\kappa_\beta)]\Big] \times X_{n+\beta,n}^{(-)}\Big[X_{n+\beta,n}^{(-)} \pm (\alpha_2/\alpha_1)^{1/2}X_{n+\beta,n}^{(+)} + \left(\frac{\alpha_4^2}{\alpha_1}\frac{\hbar q^2}{2m\omega_0}\right)^{1/2} f_{n+\beta,n}\Big]\Big\}, \quad (30)$$

where $f_0(E)$ is the Fermi distribution function and κ_β is given by

$$\kappa_\beta = (m/\hbar q)(\alpha_3-\alpha_4^2/\alpha_2)^{-1}(\omega-\beta\omega_0)-q/2. \quad (31)$$

The summations are to be performed over all β from $-n$ to infinity and all n from zero to infinity. The $\Delta n=0$ transitions are those for which $\beta=0$. The functions $f_{n+\beta,n}$ and $X_{n+\beta,n}^{(\pm)}$ can be expressed in terms of Bessel functions in the semiclassical limit.[3] The oscillatory nature of the Bessel functions is the origin of the resonances.

From Eqs. (30) and (31), one can also see that the higher transitions, $\Delta n=\pm 1$, for example, also show geometric resonances. These resonances measure *non-extremal* orbits[12] and may be useful in studies of Fermi surfaces. The $\Delta n=\beta$ transitions measure properties of electron orbits for which k_z, the component of wave vector parallel to the magnetic field, is equal to κ_β. A thorough analyses of the attentuation of helicon waves in anisotropic materials, including both the semiclassical geometric resonances and quantum effects will be presented in a later publication.

APPENDIX

The functions $f_{n'n}(\mathbf{q})$ defined by Eq. (22) can be expressed in terms of associated Laguerre polynomials. It is straightforward but rather tedious to prove that for $n' \geq n$

$$f_{n'n}(\mathbf{q}) = \left(\frac{n!}{n'!}\right)^{1/2}\exp\{-\tfrac{1}{4}(s+it)^2\}\left(\frac{s+it}{\sqrt{2}}\right)^{n'-n} \times L_n^{n'-n}\left(\frac{s^2+t^2}{2}\right). \quad (A1)$$

In this equation $L_n^{n'-n}(\xi)$ is an associated Laguerre polynomial, and the parameters s and t are defined by the relations

$$s = (\hbar\alpha_2/m\omega_0)^{1/2}\left(q_y+\frac{\alpha_4}{\alpha_2}q_z\right), \quad (A2)$$

[10] See, for example, R. Bowers, C. Legendy, and F. Rose, Phys. Rev. Letters **7**, 339 (1961); R. G. Chambers and B. K. Jones, Proc. Roy. Soc. (London) **A270**, 417 (1962); and F. Rose, M. T. Taylor, and R. Bowers, Phys. Rev. **127**, 1122 (1962).

[11] The fact that $\Delta n=0$ transitions are allowed for anisotropic materials may make it difficult to accurately locate the position of the absorption edge at $(qv_F/\omega_0)=1$ which was discussed by E. A. Stern, Phys. Rev. Letters **10**, 91 (1963).

[12] The geometric resonances in ultrasonic attenuation due to $n' \neq n$ transitions have recently been considered by Y. Eckstein (private communication). I am indebted to Dr. Eckstein for pointing out the importance of these resonances.

and

$$t=(\hbar\alpha_1/m\omega_0)^{1/2}q_x. \tag{A3}$$

It is not difficult to demonstrate that

$$f_{n'n}(-\mathbf{q})=(-1)^{n'+n}f_{n'n}(\mathbf{q}) \tag{A4}$$

and that

$$f_{nn'}(\mathbf{q})=\exp(-ist)f_{n'n}^*(-\mathbf{q}).$$

There exist some useful sum rules for the functions $f_{n'n}(\mathbf{q})$ and $X_{n'n}^{(\pm)}(\mathbf{q})$ which are similar to those given in the Appendix of Ref. 3. The reader is referred to this reference for further details.

PHYSICAL REVIEW VOLUME 135, NUMBER 1A 6 JULY 1964

Variation of Nuclear Spin Polarization Time with Excitation of Electron Resonances*

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(Received 6 February 1964)

In solid dielectric materials where nuclear and electron spins are coupled by dipolar interaction, positive and negative enhancements of the nuclear polarization are produced by inducing simultaneous electron-nuclear transitions (double-flip transitions). The nuclear polarization time τ_N is defined to be the time constant associated with the approach of the nuclear spin system to the enhanced state. We have measured the proton polarization time of several such materials and found that τ_N is less than the thermal relaxation time when double flips are induced and greater, sometimes by two orders of magnitude, when the central electron resonance is excited. The first effect is understandable from the spin polarization rate equations. One expects, qualitatively, that the second effect can be explained by a decoupling of electron and proton spins due to stirring of electron spins by the applied rf field; such an explanation is shown to be incorrect.

I. INTRODUCTION

IN 1953, Overhauser[1] proposed a method whereby the polarization of the nuclear spin states in a metal could be enhanced by saturation of the paramagnetic resonance of the conduction electron spins. It was soon pointed out by others[2] that spin polarization enhancement is not limited to metals but could be obtained whenever two spin systems are coupled and an rf magnetic field is applied at, or near, the Larmor frequency of one of them. (The term "rf" is understood to include microwave frequencies.) We are here concerned with solid dielectric materials containing paramagnetic centers in which the paramagnetic spins S are coupled to the nuclear spins I by dipolar interaction. It is well known that in such materials the nuclear spin polarization is enhanced when simultaneous electron-nuclear spin flips are induced.[3–5] These double-flip transitions are weakly forbidden and are possible because of mixing of the spin states by the dipolar interaction. When the frequency of the applied rf magnetic field is $\nu_e-\nu_N$ (ν_e and ν_N are, respectively, the electron and nuclear Larmor frequencies) the polarization is positively enhanced and when the frequency is $\nu_e+\nu_N$ the polarization enhancement is negative. The enhancement is zero when pure electronic transitions only are induced. This type of enhancement is commonly referred to as the "solid-state effect."

When enhancement is caused by exciting a double-flip transition the nuclear spin system approaches the enhanced population distribution exponentially with the characteristic time τ_N, the polarization time. Upon removal of the excitation the spin system decays to thermal equilibrium, the rate being given by the thermal relaxation time T_{1N}.

The electron spins exchange energy with the lattice directly via electron-phonon interaction, while direct contact of the nuclear spins with the lattice is very weak and energy is exchanged via the dipolar interaction with the electron spins. The rate of exchange of energy between the nuclear spins and the lattice depends on the spectral density, at frequency ν_N, of the time varying, electron-induced dipolar field at the nuclear sites. Thus, one would expect nuclear polarization and relaxation times to depend on the rate of stirring of the electron spins by the applied rf field since rapid stirring can alter the spectral density if the rf field is large enough. Qualitatively, we expect the following: (1) When pure electronic transitions are induced, the forced flipping of the electron spins will decouple them from the nuclear spins causing a de-

* This work was supported, in part, by the U. S. Air Force Office of Scientific Research, Solid State Science Division and the National Science Foundation.
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