

Modification of the Velocity of Sound in Metals by an Applied Magnetic Field*

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(Received 11 January 1963)

An analysis is given of the change in the velocity of sound as a function of an applied magnetic field. It is found that at room temperature and for ultrasonic frequencies of a few Mc/sec, the velocity of sound increases by an amount proportional to the square of the magnetic field in agreement with the experimental results. A brief discussion is given of the change in the sound velocity as a function of a magnetic field at low temperatures.

I. INTRODUCTION

MEASUREMENTS of the velocity of sound in metals in the presence of an applied magnetic field have been carried out by Alers and Fleury.¹ These measurements have been made on single crystals of Cu, Ag, Au, Al, Ta, and V at room temperature and using 10 Mc ultrasonic pulses. They have shown that the change in the velocity of sound is proportional to the square of the magnetic field \mathbf{B}_0 and depends on different geometrical arrangements in a way that will be described below. There has also been experimental work by Galkin and Koroliuk² and by Beattie, Silsbee, and Uehling³ in a few polycrystalline metals. They obtain results similar to those of reference 1.

A macroscopic theory which accounts for the main features of this phenomenon has been given by Alpher and Rubin.⁴ However, a microscopic theory seems to be desirable. The first step towards the construction of such a theory has been taken by Harrison.⁵ The model used by Harrison is that of a metal consisting of a free-electron gas and a uniform background of positively charged ions. The present work makes use of the same model but differs in that we have considered the collisions of the conduction electrons with lattice imperfections such as thermal phonons, impurity atoms, etc. Therefore, this work is directly applicable to the experimental work in which the electron relaxation time τ is much smaller than the period of the acoustic waves. Furthermore, in the limit of zero magnetic field it yields the well-known^{6,7} result for the velocity of longitudinal sound for this model. Finally, the results of this paper are applicable to both longitudinal and shear waves.

We now describe some of the experimental results of Alers and Fleury. For longitudinal waves the relative change in the velocity of sound is $(B_0^2 \sin^2 \theta / 8\pi \rho s_l^2)$, where ρ is the density of the material, s_l is the speed of

longitudinal acoustic waves, and θ is the angle between the direction of propagation of the sound wave and the magnetic field \mathbf{B}_0 . For shear waves the relative change in the velocity of sound is $(B_0^2 \cos^2 \theta / 8\pi \rho s_t^2)$ and is the same for all transverse waves independently of the direction of polarization. The quantity s_t is the speed of shear acoustic waves in the absence of the magnetic field. These results can also be obtained by a simple generalization⁸ of the theory of Alpher and Rubin. In this paper, we discuss these results under several circumstances. First, we consider the conditions applicable to the experiments of Alers and Fleury where we expect $\omega\tau \approx 10^{-6}$. Here ω is the angular frequency of the acoustic wave ($\omega = 6.28 \times 10^7 \text{ sec}^{-1}$). Ordinarily $\tau \approx 10^{-14}$ sec for a metal such as copper at room temperature. This discussion together with a few examples of calculations carried out for situations in which $\omega\tau$ is not negligible (low temperatures) is given in the next section.

II. THEORY

Let us consider an acoustic wave of angular frequency ω and wave vector \mathbf{q} whose displacement field $\xi(\mathbf{r}, t)$ at position \mathbf{r} and time t is given by

$$\xi = \xi_0 \exp(i\omega t - i\mathbf{q} \cdot \mathbf{r}). \quad (1)$$

We assume that we have n_0 conduction electrons and n_0/z positive ions per unit volume (i.e., there are z conduction electrons per atom). The equation of motion of one ion is

$$M \partial^2 \xi / \partial t^2 = C_l \nabla (\nabla \cdot \xi) - C_t \nabla \times (\nabla \times \xi) - ze\mathbf{E} - (ze/c)\mathbf{u} \times (\mathbf{B}_0 + \mathbf{B}) + \mathbf{F}_e. \quad (2)$$

Here M is the mass of the ion, C_l and C_t are elastic constants describing the interaction between the ion cores in the absence of the long-range Coulomb interactions between ions and electron, $\mathbf{u} = \partial \xi / \partial t = i\omega \xi$ is the velocity field associated with the acoustic wave, and e is the charge on the electron. The vectors \mathbf{E} and \mathbf{B} are the electric field and magnetic induction of the electromagnetic field that arises because of the passage of the acoustic wave through the metal. The average force \mathbf{F}_e owes its origin to the collisions of the electrons with lattice imperfections. This force can be described by

* S. Rodriguez, Phys. Letters 2, 271 (1962).

* Supported in part by the Advanced Research Projects Agency.

¹ G. A. Alers and P. A. Fleury, Phys. Rev. 129, 2425 (1963).

² A. A. Galkin and A. P. Koroliuk, Zh. Eksperim. i Teor. Fiz. 34, 1025 (1958) [translation: Soviet Phys.—JETP 7, 708 (1958)].

³ A. G. Beattie, H. B. Silsbee, and E. A. Uehling, Bull. Am. Phys. Soc. 7, 478 (1962).

⁴ R. A. Alpher and R. J. Rubin, J. Acoust. Soc. Am. 26, 452 (1954).

⁵ M. J. Harrison, Phys. Rev. Letters 9, 299 (1962); see also, T. Kjeldaas, Phys. Rev. 113, 1473 (1959).

⁶ D. Bohm and T. Staver, Phys. Rev. 84, 836 (1952).

⁷ J. Bardeen and D. Pines, Phys. Rev. 99, 1140 (1955).

introducing a phenomenological time τ , the average time between two successive collisions of one electron. In Eq. (2) we have assumed that the forces between the ion cores are elastically isotropic. This is certainly not the case in practice. However, this assumption has been made for the sake of simplicity and because our purpose is to investigate the effect of the conduction electrons on the propagation of acoustic waves in metals.

An expression for the force \mathbf{F}_c can be obtained as follows. If $\mathbf{j}^{(1)}(\mathbf{r}, t)$ is the electron current density within the metal at position \mathbf{r} and time t , the average velocity of the electrons is $\langle \mathbf{v} \rangle = \mathbf{j}^{(1)}/n_0 e$ so that each ion experiences an average increase in its momentum in the amount $(zm/\tau)\langle \mathbf{v} \rangle - \mathbf{u}$ per unit time. Thus,

$$\mathbf{F}_c = (mz/\tau)[\langle \mathbf{v} \rangle - \mathbf{u}] = (zm/n_0 e \tau) \mathbf{j}^{(1)} - (mz/\tau) \mathbf{u}. \quad (3)$$

In the fourth term on the right-hand side of Eq. (2), \mathbf{B} can be neglected without introducing an appreciable error. Substitution of \mathbf{F}_c given by Eq. (3) into Eq. (2) yields a relation containing the self-consistent electric field \mathbf{E} , the electron current density $\mathbf{j}^{(1)}$, and the displacement ξ . We must now establish relations connecting \mathbf{E} , $\mathbf{j}^{(1)}$, and ξ . One relation is provided by Maxwell's equations connecting the total current density

$$\mathbf{j} = \mathbf{j}^{(1)} - n_0 e \mathbf{u}, \quad (4)$$

to \mathbf{E} by an equation of the form⁹

$$\mathbf{j} = \mathbf{\Gamma} \cdot \mathbf{E}. \quad (5)$$

Here $\mathbf{\Gamma}$ is the tensor

$$\mathbf{\Gamma} = \frac{ic^2 q^2}{4\pi\omega} \left[1 - \frac{\omega^2}{c^2 q^2} \right] \mathbf{I} - \frac{ic^2}{4\pi\omega} \mathbf{q}\mathbf{q}, \quad (6)$$

where \mathbf{I} is the unit tensor. A second relation which is required to obtain a complete solution is the so-called constitutive equation which gives the electron current density in terms of the field \mathbf{E} . This current density has components originating from three different causes. The first component is the current caused by the direct action of the electric field \mathbf{E} , the second comes about because the electrons, upon being scattered by the lattice, retain a drift velocity \mathbf{u} , and the third is a diffusion current that takes place because the dilations and contractions of the lattice alter locally the position of the Fermi level giving rise to currents from regions of higher concentration to regions of lower concentration. The constitutive equation is found by standard arguments of transport theory which we shall briefly discuss.

Let us designate the probability of finding an electron in a state described by the wave vector \mathbf{k} and at position \mathbf{r} and time t by $f(\mathbf{k}, \mathbf{r}, t)$. In equilibrium $f(\mathbf{k}, \mathbf{r}, t)$ reduces to the Fermi-Dirac distribution $f_0(\mathbf{k})$. We assume that

in the presence of the acoustic wave f differs from f_0 by an amount $f_1(\mathbf{k}, \mathbf{r}, t)$ which is small as compared to $f_0(\mathbf{k})$ for each value of \mathbf{k} :

$$f(\mathbf{k}, \mathbf{r}, t) = f_0(\mathbf{k}) + f_1(\mathbf{k}, \mathbf{r}, t). \quad (7)$$

Furthermore, f_1 varies as $\exp(i\omega t - i\mathbf{q} \cdot \mathbf{r})$. The present formulation is valid if the de Broglie wavelength of an electron at the top of the Fermi distribution is much smaller than the acoustic wavelength. This is satisfied if $\omega \ll msv_0/\hbar$, where m is the mass of the electron, s is the velocity of sound, and v_0 is the Fermi velocity (for a typical metal $msv_0/\hbar \approx 5 \times 10^{13} \text{ sec}^{-1}$). The distribution function must satisfy the Boltzmann transport equation for the motion of the electron gas in the presence of the fields \mathbf{B}_0 and \mathbf{E} . In place of the collision term we use $-(f - \bar{f}_0)/\tau$ where \bar{f}_0 is not equal to the equilibrium distribution f_0 but rather, to the local equilibrium distribution. \bar{f}_0 differs from f_0 in two respects. First, \bar{f}_0 depends on the kinetic energy of the electron measured in a coordinate system moving with the velocity \mathbf{u} rather than on the kinetic energy with respect to the laboratory system. Second, the Fermi energy associated with \bar{f}_0 is the local Fermi energy $\zeta(\mathbf{r}, t)$ rather than its equilibrium value ζ_0 . Let $n(\mathbf{r}, t) = n_0 + n_1(\mathbf{r}, t)$ be the electron concentration at \mathbf{r} and t . Then, since $n_1 \ll n_0$ we have

$$\zeta(\mathbf{r}, t) = \zeta_0 + n_1/g(\zeta_0), \quad (8)$$

where $g(\epsilon) = 3n_0 \epsilon^{1/2}/2\zeta_0^{3/2}$ is the density of electronic states per unit volume and per unit energy range at ϵ (ϵ is the kinetic energy of an electron). Since the relative kinetic energy of an electron in the state \mathbf{k} is $\epsilon_k - m\mathbf{v}_k \cdot \mathbf{u}$, where \mathbf{v}_k is the electron velocity, we have

$$f_0(\mathbf{k}, \mathbf{r}, t) = f_0(\mathbf{k}) - [m\mathbf{v}_k \cdot \mathbf{u} + n_1/g(\zeta_0)] \frac{\partial f_0}{\partial \epsilon}. \quad (9)$$

From this result, it follows (even in the case in which a magnetic field is present) that

$$\mathbf{j}^{(1)} = \boldsymbol{\sigma} \cdot \boldsymbol{\mathcal{E}} - e\mathbf{D} \cdot \nabla n. \quad (10)$$

In Eq. (10) $\boldsymbol{\sigma}$ is the electrical conductivity tensor, \mathbf{D} is the diffusion tensor given by

$$\mathbf{D} = \boldsymbol{\sigma}/e^2 g(\zeta_0)(1 + i\omega\tau), \quad (11)$$

and

$$\boldsymbol{\mathcal{E}} = \mathbf{E} + (m\mathbf{u}/e\tau). \quad (12)$$

These results have been discussed in detail in reference 9. We are now in a position to solve for \mathbf{E} and $\mathbf{j}^{(1)}$ in terms of the displacement field. It is convenient to use the fact that the operations $\partial/\partial t$ and ∇ are equivalent to multiplication by $i\omega$ and $-i\mathbf{q}$, respectively, because all variable quantities behave in the same fashion as ξ . Equation (2) can be rewritten in the form

$$M\omega^2 \xi = (C_l - C_t)\mathbf{q}\mathbf{q} \cdot \xi + C_t q^2 \xi + (ze/\sigma_0)(\mathbf{I}\sigma_0 - \mathbf{\Gamma}) \cdot \mathbf{E} + (zei\omega/c)\xi \times \mathbf{B}_0, \quad (13)$$

⁹ M. H. Cohen, M. J. Harrison, and W. A. Harrison, Phys. Rev. **117**, 937 (1960). The reader is referred to this work for a more detailed discussion of Eqs. (4)–(12).

where

$$\mathbf{E} = [eg(\zeta_0)(1+i\omega\tau)]^{-1}(\mathbf{I}-\mathbf{R}\cdot\mathbf{\Gamma})^{-1}\cdot\nabla n - (n_0e i\omega/\sigma_0)(\mathbf{I}-\mathbf{R}\cdot\mathbf{\Gamma})^{-1}(\mathbf{I}-\sigma_0\mathbf{R})\cdot\xi. \quad (14)$$

The tensor \mathbf{R} is the resistivity tensor, i.e., the reciprocal of σ and $\sigma_0 = n_0e^2\tau/m$ is the ordinary dc conductivity. The gradient of the electron concentration can be obtained from the equation of continuity for the electron charge density. To obtain ∇n we neglect the displacement current. This approximation is valid at all but the highest ultrasonic frequencies and is equivalent to the assumption of quasi-neutrality. Then

$$\nabla n = n_0\mathbf{q}\cdot\xi. \quad (15)$$

Equations (13)–(15) permit us to obtain a secular equation giving the complex angular frequencies of acoustic waves in terms of \mathbf{q} , \mathbf{B}_0 and the parameters that characterize our model of the metal.

We shall discuss first the experimental situation obtained in the experiments of Alers and Fleury. Under the conditions of their work both $\omega\tau$ and $\omega_c\tau$ (where $\omega_c = -eB_0/mc$ is the cyclotron frequency) are negligible as compared to unity. Also $ql \ll 1$, where $l = v_0\tau$ is the electron mean free path. Under such conditions the resistivity tensor acquires a very simple form that can be obtained from the solution of the transport equation or from simpler considerations.⁸ We shall assume that \mathbf{q} is directed along the z axis of a Cartesian coordinate system and that the magnetic field $\mathbf{B}_0 = (0, B_0 \sin\theta, B_0 \cos\theta)$ lies in the y - z plane and makes an angle θ with \mathbf{q} . The components of the tensors $\mathbf{\Gamma}$ and \mathbf{R} are given, under these conditions, by

$$\Gamma_{xx} = \Gamma_{yy} = i\beta\sigma_0, \quad (16)$$

$$\Gamma_{zz} = -i\omega/4\pi, \quad (17)$$

$$R_{xx} = R_{yy} = R_{zz} = 1/\sigma_0, \quad (18)$$

$$R_{xy} = -R_{yx} = \omega_c\tau \cos\theta/\sigma_0, \quad (19)$$

$$R_{xz} = -R_{zx} = -\omega_c\tau \sin\theta/\sigma_0, \quad (20)$$

and

$$R_{yz} = R_{zy} = 0. \quad (21)$$

The parameter β is defined by

$$\beta = c^2q^2/4\pi\omega\sigma_0. \quad (22)$$

We are now in a position to obtain the components of the electric field \mathbf{E} by calculating the tensors $(\mathbf{I}-\mathbf{R}\cdot\mathbf{\Gamma})^{-1}$ and $(\mathbf{I}-\mathbf{R}\cdot\mathbf{\Gamma})^{-1}(\mathbf{I}-\sigma_0\mathbf{R})$. Since we are only interested in obtaining the first-order correction to the speed of sound, these tensors are calculated keeping terms up to $(\omega_c\tau)^2$ along the diagonals and up to $\omega_c\tau$ off the diagonals. This is permissible since we assumed $\omega_c\tau \ll 1$. The parameter β can be of the order of unity or less. In the absence of a magnetic field the acoustic waves which one obtains are purely longi-

tudinal or transverse. Strictly speaking this is no longer true when the magnetic field \mathbf{B}_0 is present. However, it is interesting to notice that under the approximation given here (i.e., keeping terms in the secular equation up to second order in $\omega_c\tau$) the acoustic waves are still either purely longitudinal or purely transverse. The calculations are rather simple so that they will be omitted. The relative change in the speed of shear waves is

$$\Delta s/s_l = B_0^2 \cos^2\theta/8\pi\rho s_l^2(1+\beta^2). \quad (23)$$

For longitudinal waves we obtain

$$\Delta s/s_l = B_0^2 \sin^2\theta/8\pi\rho s_l^2(1+\beta^2). \quad (24)$$

We remark that the value one obtains for s_l is the same one as that given by Bohm and Staver. The results given by (23) and (24) are in good agreement with the experimental data of reference 1.

We now obtain general formulas for the change in the sound velocity for propagation both along the direction of the magnetic field \mathbf{B}_0 and at right angles to it. In each case we orient the Cartesian coordinate system so that \mathbf{B}_0 points along the z axis. We first consider the case in which \mathbf{q} is parallel to \mathbf{B}_0 . In this case the conductivity tensor has been given by several authors.⁹ We have $\Gamma_{xx} = \Gamma_{yy} = i\beta\sigma_0$ and $\sigma_{xx} = \sigma_{yy}$ and $\sigma_{xy} = -\sigma_{yx}$. It is convenient to rewrite the general equations (13) and (14) using $\xi_{\pm} = \xi_x \pm i\xi_y$ and $\sigma_{\pm} = \sigma_{xx} \mp i\sigma_{xy}$. We find

$$\omega_{\pm}^2 = (C_l/M)q^2 \pm \frac{ze\omega B_0}{Mc} + \frac{zmi\omega}{M\tau} \frac{(1-i\beta)(\sigma_0 R_{\pm} - 1)}{1-i\beta\sigma_0 R_{\pm}}, \quad (25)$$

and

$$\omega^2 = \frac{zm}{3M} \frac{q^2 v_0^2}{1+\omega^2\tau^2} + \frac{zmi\omega}{M\tau} \left[\sigma_0 R_{zz} - 1 - \frac{1}{3} \frac{(ql)^2}{1+\omega^2\tau^2} \right]. \quad (26)$$

Equation (25) refers to the two shear waves in ξ_{\pm} and (26) to the longitudinal wave. In (26) the quantity $(C_l/M)q^2$ has been neglected as compared with $zmq^2v_0^2/3M$. The quantities R_{\pm} and R_{zz} are the reciprocals of σ_{\pm} and σ_{zz} , respectively. Within the framework of the semiclassical transport theory R_{zz} turns out to be independent of the field B_0 . The discussion of quantum effects in this connection has been given by Quinn and Rodriguez.¹⁰ It seems useful to make the remark that from Eqs. (25) and (26) we find not only the sound velocity as a function of B_0 but also the coefficient of ultrasonic attenuation $\gamma = 2\omega_2/s$, where ω_2 is the imaginary part of ω as given by either (25) or (26) and s is the appropriate sound velocity.

When $ql \ll 1$, we find

$$\sigma_0 R_{\pm} = 1 + i\omega\tau \mp i\omega_c\tau. \quad (27)$$

In this expression $\omega_c\tau$, however, can be quite large.

¹⁰ J. J. Quinn and S. Rodriguez, Phys. Rev. Letters 9, 145 (1962).

Then after assuming that $|\beta\sigma_0 R_{\pm}| \ll 1$,

$$\left(\frac{\omega_{\pm}}{q}\right)^2 = \frac{C_t}{M} + \frac{B_0^2}{4\pi\rho} \left(1 \mp \frac{\omega}{\omega_c}\right)^2. \tag{28}$$

This equation is implicit in the work of Kjeldaas (see reference 5). This implies that, for shear wave propagating along the direction of a magnetic field there is a different sound velocity for the right and left circularly polarized components. This gives rise to a rotation of the plane of polarization proportional to B_0 as the wave progresses within the metal. However, the angle of rotation turns out to be of the order of 10^{-10} deg per cm of path and per G of applied field. This is negligible under almost any circumstance.

We now turn our attention to the other geometrical arrangement of interest. This is the one in which \mathbf{q} is perpendicular to \mathbf{B}_0 . Now we choose the y axis parallel to \mathbf{q} and the z axis parallel to \mathbf{B}_0 as before. The components of the resistivity tensor are now

$$R_{xx} = \frac{\sigma_{yy}}{\sigma_{xx}\sigma_{yy} + \sigma_{xy}^2}, \tag{29}$$

$$R_{yy} = \frac{\sigma_{xx}}{\sigma_{xx}\sigma_{yy} + \sigma_{xy}^2}, \tag{30}$$

$$R_{xy} = -R_{yx} = -\frac{\sigma_{xy}}{\sigma_{xx}\sigma_{yy} + \sigma_{xy}^2}, \tag{31}$$

and

$$R_{zz} = 1/\sigma_{zz}. \tag{32}$$

Using Eqs. (13) and (14) we obtain

$$\omega^2 \xi_x = A_{xx} \xi_x + A_{xy} \xi_y, \tag{33}$$

$$\omega^2 \xi_y = A_{yx} \xi_x + A_{yy} \xi_y, \tag{34}$$

and

$$\omega^2 \xi_z = A_{zz} \xi_z, \tag{35}$$

where

$$A_{xx} = \frac{C_t}{M} q^2 + \frac{zmi\omega}{M\tau} \frac{(\sigma_0 R_{xx} - 1)(1 - i\beta)}{1 - i\beta\sigma_0 R_{xx}}, \tag{36}$$

$$A_{xy} = -A_{yx} = \frac{zmi\omega}{M\tau} \left(\frac{(1 - i\beta)\sigma_0 R_{xy}}{1 - i\beta\sigma_0 R_{xx}} - \omega_c \tau \right), \tag{37}$$

$$A_{yy} = \frac{C_t}{M} q^2 + \frac{zmq^2 v_0^2}{3M(1 + \omega^2 \tau^2)} + \frac{zmi\omega}{M\tau} \times \left(\sigma_0 R_{yy} - 1 - \frac{i\beta(\sigma_0 R_{xy})^2}{1 - i\beta\sigma_0 R_{xx}} - \frac{(ql)^2}{3(1 + \omega^2 \tau^2)} \right), \tag{38}$$

and

$$A_{zz} = \frac{C_t}{M} q^2 + \frac{zmi\omega}{M\tau} \frac{(\sigma_0 R_{zz} - 1)(1 - i\beta)}{1 - i\beta\sigma_0 R_{zz}}. \tag{39}$$

Equations (33)–(35) show that in this geometry a wave

polarized along the z axis is purely transverse. However, the normal modes that arise from longitudinal waves and from shear waves polarized along the x axis are mixed. For the wave that is approximately longitudinal we have

$$\omega^2 = A_{yy} - A_{xy}^2 / (s_l^2 - s_t^2) q^2, \tag{40}$$

while for the waves that are almost transverse and polarized along the x axis

$$\omega^2 = A_{xx} + A_{xy}^2 / (s_l^2 - s_t^2) q^2. \tag{41}$$

In these equations we have assumed that $(s_l^2 - s_t^2) \times q^2 \gg A_{xy}^2$ which is certainly the case except for magnetic fields for which quantum effects play an important role. An obvious approximation has also been made in the denominators of the second terms of Eqs. (40) and (41). Inspection of Eqs. (36)–(39) shows that the velocity of sound is not, in general, a monotonic function of the magnetic field. In fact, the velocity of sound experiences oscillations of the same nature as those found in reference 9 for the coefficient of ultrasonic attenuation. To exhibit this behavior we shall discuss the situation that obtains at low temperatures for metals of high purity in high magnetic fields. More specifically, we study the case in which $\omega_c \tau \gg 1$ and $\beta \ll \omega \tau < 1$. The second inequality requires that the dc electrical conductivity σ_0 be much larger than $\omega_p c q / 4\pi\omega$, where ω_p is the plasma frequency of the conduction electrons of the metal. A more detailed analysis is reserved for a future publication.

Using these approximations and the results of Cohen *et al.*⁹ we obtain the relations

$$A_{xx} = \frac{C_t}{M} q^2 + \frac{zmi\omega}{M\tau} S_{xx} - \frac{zm\omega^2}{M} (1 + S_{xx}), \tag{42}$$

$$A_{yy} = \frac{C_t}{M} q^2 + \frac{zmq^2 v_0^2}{3M} + \frac{zmi\omega}{M\tau} (S_{yy} - 1) + \frac{zm\omega^2}{M} S_{yy}, \tag{43}$$

$$A_{xy} = -\frac{zmi\omega}{M} (\omega_c + qv_0 S_{xy}), \tag{44}$$

and

$$A_{zz} = \frac{C_t}{M} q^2 + \frac{zmi\omega}{M\tau} S_{zz} - \frac{zm\omega^2}{M} (1 + S_{zz}). \tag{45}$$

In these equations S_{xx} , S_{yy} , S_{xy} , and S_{zz} (see reference 9) are given by

$$S_{xx} = \frac{1}{3} [s_0 + (g_0'/2)^2 / (1 - g_0)]^{-1} - 1, \tag{46}$$

$$S_{yy} = \frac{q^2 l^2}{3(1 + \omega^2 \tau^2)} \left[\frac{s_0}{(1 - g_0)s_0 + (g_0'/2)^2} - 1 \right], \tag{47}$$

$$S_{xy} = \frac{(g_0'/2)}{3[s_0(1 - g_0) + (g_0'/2)^2]}, \tag{48}$$

and

$$S_{zz} = (1/3r_0) - 1, \tag{49}$$

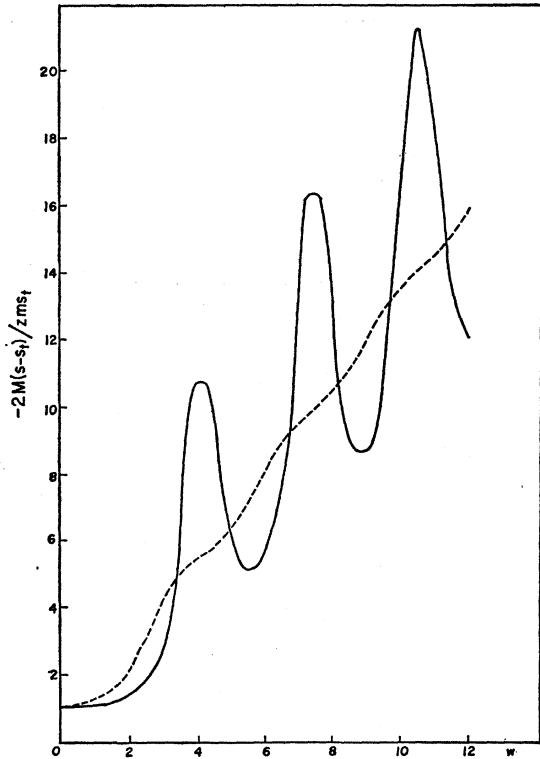


FIG. 1. Relative change in the velocity of shear waves as a function of $w = qv_0/\omega_c$. The wave propagates along the y axis and the magnetic field is parallel to the z axis. The full line corresponds to a shear wave polarized along the x direction and the broken line corresponds to polarization parallel to the magnetic field.

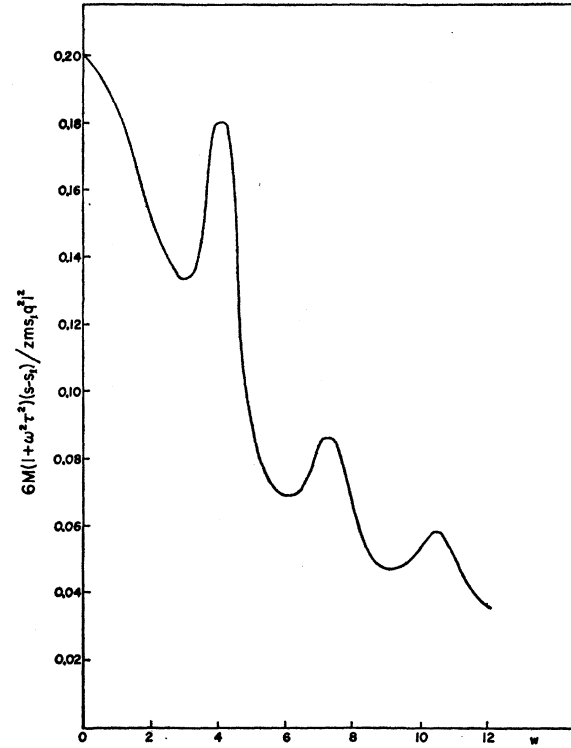


FIG. 2. Relative change in the velocity of a longitudinal sound wave propagating in a direction perpendicular to the magnetic field as a function of w .

with the functions g_0 , s_0 , and r_0 defined by means of the relations

$$g_0(w) = \int_0^{\pi/2} J_0^2(w \sin\theta) \sin\theta d\theta, \quad (50)$$

$$s_0(w) = \int_0^{\pi/2} [J_0'(w \sin\theta)]^2 \sin^3\theta d\theta, \quad (51)$$

and

$$r_0(w) = \int_0^{\pi/2} J_0^2(w \sin\theta) \cos^2\theta \sin\theta d\theta. \quad (52)$$

Here

$$w = qv_0/\omega_c, \quad (53)$$

and J_0 is the Bessel function of order zero. It is useful to remark that for large magnetic fields ($w \ll 1$) the expression for A_{xy} approaches zero. In fact, using Eqs. (48), (51), and (52), we find that for $w \ll 1$, $S_{xy} = -\omega_c/qv_0$. Now, since in Eqs. (40) and (41), A_{xy} appears to the second power it is a simple matter to see that its contribution to the change in the velocity of sound is negligible. In fact, the term $A_{xy}^2/(s_l^2 - s_t^2)q^2$ can be neglected in comparison with contributions of the form $zm\omega^2(1+S_{xx})/M$ and similar ones because

m/M is of the order of 10^{-4} or less and A_{xy}^2 contains this ratio to the second power. We can now calculate the relative change in the sound velocity under these conditions. We find

$$(s - s_t)/s_t = -zm(1+S_{xx})/2M, \quad (54)$$

for shear waves polarized along the x axis. For longitudinal waves we obtain

$$(s - s_l)/s_l = zmS_{yy}/2M, \quad (55)$$

while the relative change in the velocity of sound for shear wave polarized along the z axis is

$$(s - s_t)/s_t = -zm(1+S_{zz})/2M. \quad (56)$$

These changes in velocity are of the order of 10 ppm and can be detected experimentally.¹ In Figs. 1 and 2 we give graphs of the relative change of the velocity of sound as a function of $w = qv_0/\omega_c$. These graphs were drawn by calculating S_{xx} , S_{yy} , and S_{zz} using the values of the functions g_0 , s_0 , and r_0 given in reference 9.

It is possible to generalize this treatment to take into account quantum effects in the variation of the velocity of sound with a magnetic field. This can be easily accomplished using the results of Quinn and Rodriguez.¹¹

¹¹ J. J. Quinn and S. Rodriguez, Phys. Rev. 128, 2487, 2494 (1962).

This effect has been observed in bismuth by Mavroides *et al.*¹² However, the theory given here is not applicable to semimetals in the present form.

¹² J. G. Mavroides, B. Lax, K. J. Button, and Y. Shapira, Phys. Rev. Letters **9**, 451 (1962).

ACKNOWLEDGMENTS

The author wishes to thank Dr. G. A. Alers for communicating his results prior to publication. He is also indebted to Dr. M. J. Harrison, Dr. J. J. Quinn, and Dr. A. W. Overhauser for stimulating discussions.

Spin Wave Spectra of Magnetite

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(Received 18 January 1963)

Expressions for spin wave energies in normal and inverse spinel have been obtained which are exact to all orders in the spin wave momentum. These dispersion curves have been compared with existing experimental data on magnetite and good agreement found if the principal exchange interaction is taken to be 2.4×10^{-3} eV. The surprisingly small range of validity of the usual k^2 approximation is pointed out and possible effects of the deviation from k^2 behavior on the magnetic part of the heat capacity are discussed. The small (like-like) exchange interactions have been included (also to all orders in the spin-wave momentum) with the most important result that agreement with the experimental dispersion curves is improved.

INTRODUCTION

MAGNETITE (Fe_3O_4) is the simplest of the so-called ferrites,¹ compounds of the form $X^{2+}(Y^{3+})_2O_4$ crystallizing in the spinel structure, (space-group $Fd\bar{3}m-O_h^7$). This structure is basically cubic having in a unit cell sixteen octahedral (*B*) sites and eight tetrahedral (*A*) sites. At normal temperatures magnetite has the inverse spinel structure in which the *A* sites are occupied by ferric ions and the remaining ferric and ferrous ions are distributed over the *B* sites. As the temperature is lowered below 120°K many magnetic and thermal properties undergo a sudden change. There is also a sharp drop in the electrical conductivity. This transformation was ascribed by Verwey² to an ordering of the ferric and ferrous ions on the *B* sites into alternate planes perpendicular to the *C* axis producing net orthorhombic symmetry. This proposal has been confirmed by neutron diffraction measurements³ and recently by Mössbauer absorption measurements.⁴ A sketch of the unit cell below the transition temperature is shown in Fig. 1.

There have been a number of calculations of the spin wave spectra in the normal and inverse spinel structure.⁵⁻⁸ Most results indicate a quadratic acoustic

branch. A linear behavior was found by Vonsovskii and Seidov, but this work has been criticized by Kaplan and Kowalewski. (The latter also points out an error in Kaplan's calculation.) Apparently the only calculation for the ordered inverse spinel structure has been made by Kouvel.⁹

The ordered structure may be considered to consist of six interpenetrating face-centered cubic lattices, two consisting of *A* sites and four of *B* sites. Thus, the spin wave spectrum will have six branches. Kouvel assumed that the *z* axis was the single anisotropy direction and that at 0°K the *A* spins were down and the *B* spins up. Assuming only nearest-neighbor *AA*, *BB*, and *AB* exchange integrals he set up the (sixth degree) secular equation for the frequencies and succeeded in finding the

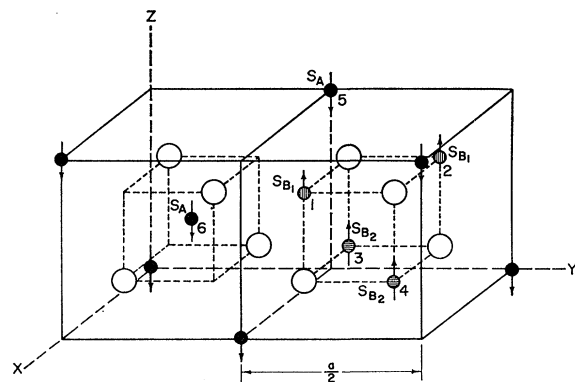


FIG. 1. One quarter of the unit cell for ordered inverse spinel. One cation site of each variety is labeled. The open circles represent oxygen sites.

¹ J. Smith and H. P. J. Wijn, *Ferrites* (John Wiley & Sons, Inc., New York, 1959).

² E. J. W. Verwey and E. L. Heilmann, J. Chem. Phys. **15**, 174 (1947).

³ W. C. Hamilton, Phys. Rev. **110**, 1050 (1958).

⁴ R. Bauminger, S. G. Cohen, A. Marinn, and E. Segal, Phys. Rev. **122**, 1447 (1961).

⁵ H. Kaplan, Phys. Rev. **86**, 121 (1952).

⁶ S. V. Vonsovskii, Y. M. Seidov, Izv. Akad. Nauk. SSSR **18**, 319 (1954) (translation available through Columbia Technical translations).

⁷ T. A. Kaplan, Phys. Rev. **109**, 782 (1958).

⁸ L. Kowalewski, Acta. Phys. Polon. **20**, 675 (1961).

⁹ J. S. Kouvel, Technical Report 210, Cruft Laboratory, Harvard, 1955 (unpublished).