Modification of the Velocity of Sound in Metals by Magnetic Fields

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The effect of a high magnetic field on the velocity of sound in single crystals of Cu, Ag, Au, Al, Ta, and V was measured using a 10-Mc/sec ultrasonic pulse technique. In agreement with theory, the velocity was found to increase as the square of the applied field and the dependence on the angle between the propagation direction and the applied field was also verified. Quantitative agreement with the macroscopic theory was excellent in the case of high conductivity metals but a slight disagreement was found at lower conductivities. Investigation of the effect at 4.2°K in extremely high purity copper where the macroscopic theory should no longer apply showed that for the field in certain crystallographic directions the velocity of sound no longer varied as the square of the magnetic field but increased linearly with field. These directions appear to correspond to some of the "open orbit" directions determined from magnetoresistance studies.

INTRODUCTION

RECENTLY there has been a renewal of theoretical interest in the very small change of the velocity of sound in metals produced by a static magnetic field. Rodriguez[1] has generalized the original macroscopic theory of Alfer and Rubin[2] and Harrison[3] has proposed a microscopic theory. All of these theories predict that the velocity of sound increases as the square of the applied magnetic field. The experiments of Galkin and Koroliuk[4] and of Beattie, Silsbee, and Uehling[5] have verified this field dependence in a few polycrystalline metals. However, if the effect is to show up any characteristics of the Fermi surface, experiments on single crystals must be carried out. It is the purpose of this paper to present the results of a series of measurements on single crystals of various purities and at a variety of temperatures in order to learn the conditions under which the present theories prove inadequate and to provide some guide lines for further theoretical development.

MACROSCOPIC THEORY

According to the theory of Alfer and Rubin[2] the effect arises from the fact that a sound wave forces a mechanical motion on the charged particles in a conducting medium. In the presence of an external magnetic field these moving charges are deflected and a transverse current analogous to a Hall current is set up. The interaction of this current with the external field produces a force which adds to the ordinary elastic forces existing in the medium. Since the sound wave is periodic in time, the Hall current is time dependent and thus magnetic and electric fields are induced in accordance with Maxwell's equations. Using $\mathbf{j}$ to represent the current, $\mathbf{e}$ the induced electric field, and $\mathbf{h}$ the induced magnetic field, one can immediately write down the relations (in cgs units)

$$\mathbf{j}=\sigma\mathbf{e}+(\sigma\mu/c)[\mathbf{v}\times(\mathbf{H}+\mathbf{h})], \tag{1}$$

$$\text{curl }\mathbf{e}=-(\mu/c)(\partial\mathbf{h}/\partial t), \tag{2}$$

$$\text{curl }(\mathbf{H}+\mathbf{h})=(4\pi/c)\mathbf{j}, \tag{3}$$

in which σ is the conductivity of the material, H the large external applied field, and $\mathbf{v}$ the instantaneous velocity of the charges. These equations are valid in a metal only so long as the electrons are tightly coupled to the sound wave. This requires that the mean free path l of the electrons be short compared to the acoustic wave length λ. Furthermore, Alfer and Rubin set $\mathbf{v}$ in Eq. (1) equal to the time derivative of the material displacement. This is acceptable only if the component of velocity produced by the magnetic field in the direction of $\mathbf{v}\times\mathbf{H}$ is small compared to $\mathbf{v}$. It can easily be shown that this condition leads to requiring $\omega_c\tau\ll1$, where ω_c is the cyclotron frequency eH/mc and τ is the mean time between collisions.

Under these two approximations ($\omega_c\tau\ll1$ and $\lambda\gg l$), the force exerted on the current $\mathbf{j}$ by the applied field makes the equation of motion governing sound wave propagation in an isotropic material become:

$$\rho\frac{\partial^2\mathbf{s}}{\partial t^2}=\frac{E}{2(1+\nu)}\left[\nabla^2\mathbf{s}+\frac{1}{1-2\nu}\nabla(\nabla\cdot\mathbf{s})\right]+\frac{\mu}{c}[\mathbf{j}\times(\mathbf{H}+\mathbf{h})], \tag{4}$$

where $\mathbf{s}$ is the medium displacement and ρ, E, and ν are the mass density, Young's modulus, and Poisson's ratio of the medium, respectively. Using Eqs. 1, 2, and 3 to eliminate the variables $\mathbf{e}$ and $\mathbf{j}$, and by letting $d\mathbf{s}/dt=\mathbf{v}$, it can be shown that Eq. (4) leads to a velocity for longitudinal sound waves given by

$$C_l=C_{0l}\left[1+\frac{\mu H^2\sin^2\theta}{8\pi\rho C_{0l}^2(1+4\pi^4\delta^4/\lambda^4)}\right], \tag{5}$$

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

[2] R. A. Alfer and R. J. Rubin, J. Acoust. Soc. Am. **26**, 452 (1954).

[3] M. J. Harrison, Bull. Am. Phys. Soc. **6**, 438 (1961); Phys. Rev. Letters **9**, 299 (1962).

[4] A. A. Galkin and A. P. Koroliuk, Soviet Phys.—JETP **7**, 708 (1958).

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

and for shear waves

$$C_s = C_{0s}\left[1+\frac{\mu H^2\cos^2\theta}{8\pi\rho C_{0s}^2(1+4\pi^4\delta^4/\lambda^4)}\right], \tag{6}$$

in which θ is the angle between the propagation direction and the field. The quantity δ is the skin depth that an electromagnetic wave with the same frequency as the sound wave would have in the material. It is related to the conductivity by

$$\delta = (c^2/2\pi\omega\sigma\mu)^{1/2}. \tag{7}$$

It is interesting to note that the magnitude of the effect is determined primarily by the ratio of the magnetic field energy density to a mechanical energy density expressed by the elastic modulus ρC^2. The fact that the material is a conductor enters the result only through a term which makes a negligible contribution in good conductors.

It is also interesting that the angular dependence for shear waves concerns the propagation direction and not the direction of particle motion even though it is the latter direction which determines the induced currents. This result comes about from the algebra of the differential operators in the equations.

EXPERIMENTAL TECHNIQUE

It can be seen from Eqs. (5) and (6) that the application of a 10-kOe magnetic field may be expected to change the sound velocity by only a few parts in a million. Such a change is easily detected and measured by the ultrasonic "sing-around" system developed to a high degree in this laboratory by Forgacs.[6] In this apparatus, a 10-Mc/sec acoustic pulse is sent through the sample in such a way that the transit time becomes the period of oscillation of an oscillator. Changes in the sound velocity produce changes in the transit time which in turn are measured as changes in the oscillator frequency.

The most serious problem associated with the use of this system arises from the temperature dependence of the velocity of sound. For most metals, the temperature coefficient of sound velocity is of the order of several hundred parts per million (ppm) per degree Kelvin. Thus, measuring a one-ppm effect requires that the specimen temperature must not change by more than 0.001°K during the measurement. Several schemes of temperature control were attempted with no success. The final arrangement involved simply allowing the temperature to drift and correcting the measurements accordingly. By simply wrapping the specimen and holder in cotton and by measuring the velocity change produced by turning the magnetic field from zero to some predetermined value, it was found possible to make the correction for temperature drift negligibly small. Using this technique the coefficient of the H^2 term in Eqs. (5) and (6), hereafter referred to as γ, could be measured to $\pm 0.1\times10^{-14}$ Oe^{-2} or about $\pm5\%$.

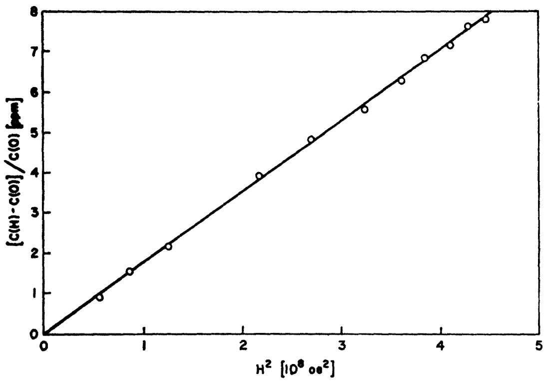

FIG. 1. Variation of the velocity of a longitudinal sound wave in gold as a function of the applied magnetic field. The solid line is predicted by the theory. Propagation direction is [110].

The magnetic field was supplied by an ADL Electromagnet operated with 5-in.-diam pole pieces separated by a 2-in. gap. With this arrangement, fields up to 21 kOe could be obtained. The field was measured with a Rawson rotating-component gaussmeter. Because the metal specimens were all smaller than a one-inch cube, no trouble with magnetic field inhomogeneity was encountered.

Since the purpose of the experiment was to find how well the macroscopic theory could be applied to real crystalline metals, all the specimens used were single crystals oriented in such a way that the sound traveled along principal crystallographic directions and, hence, were pure modes. Both longitudinal and shear type waves were used. The specimen holders were designed to hold the acoustic propagation direction fixed in the laboratory while the magnetic field direction could be

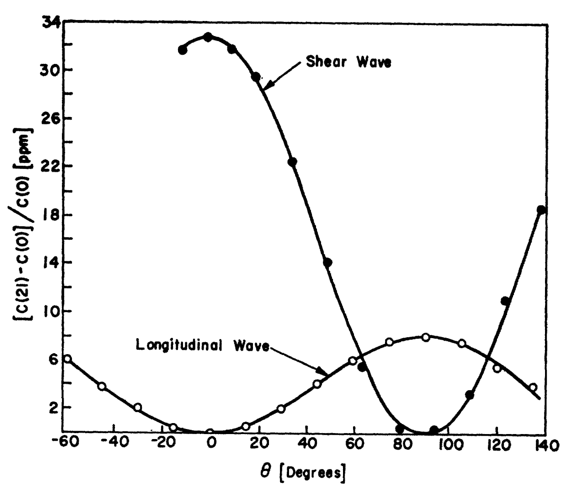

FIG. 2. Variation of the change in sound velocity accompanying the application of a 21-kOe magnetic field with the angle θ between the field direction and the [110] propagation direction. The solid lines are the angular dependence predicted by the theory.

[6] R. L. Forgacs, IRE. Trans. Instr. 9, 359 (1960).

TABLE I. Numerical results of the effect of a magnetic field on the velocity of sound. **K** is the propagation vector, **u** the polarization vector, and C_0 the velocity of the sound wave. The experimental value for the coefficient of the H^2 term in Eq. (5) or (6) is represented by γ_e and the theoretical value by γ_c. The electrical conductivity of the metal sample is given by σ.

Metal	**K**	**u**	C_0 (10^5 cm/sec)	σ (10^6 mho/cm)	$1/8\pi\rho C_0^2$ (10^{-14} Oe^{-2})	γ_c (10^{-14} Oe^{-2})	γ_e (10^{-14} Oe^{-2})	γ_c-γ_e (10^{-14} Oe^{-2})
Cu	[110]	[110]	4.97	0.591	1.79	1.79	1.8	0
	[111]	[111]	5.16		1.67	1.67	1.6	+0.1
	[110]	[001]	2.91		5.25	5.25	5.4	−0.1
	[110]	[$\bar{1}$10]	1.62		16.8	15.04	14.3	+0.7
Au	[110]	[110]	3.37	0.41	1.80	1.78	1.8	0
	[110]	[001]	1.47		9.44	7.19	6.8	+0.4
	[110]	[$\bar{1}$10]	0.87		27.3	7.56	8.1	−0.5
Ag	[100]	[100]	3.44	0.682	3.20	3.19	3.2	0
Al	[110]	[110]	6.46	0.354	3.50	3.49	3.2	+0.3
Ta	[100]	[100]	4.00	0.064	1.48	1.19	1.1	+0.1
	[100]	[001]	2.22	0.04	4.85	1.39	1.6	−0.2
V	[110]	[110]	6.65		1.84	1.72	1.4	+0.3
	[110]	[001]	2.66		9.34	3.32	3.3	0
	[110]	[$\bar{1}$10]	3.01		7.28	2.84	1.8	+1.0

rotated either about the propagation direction or in a plane containing it.

For tests below room temperature, the crystal and its support were surrounded by liquid nitrogen or liquid helium contained in a metal Dewar flask designed to fit inside the 2-in. magnetic gap and still accomodate the specimen holder.

RESULTS

The theoretical result expressed in Eq. (5) predicts that the velocity of a longitudinal sound wave should increase as the square of the magnetic field. Thus a plot of the relative change in velocity vs H^2 should be a straight line whose slope is given by the reciprocal of $8\pi\rho C_0^2$ if the field is perpendicular to the propagation direction and $\delta \ll \lambda$. Figure 1 shows an example of data obtained under these conditions on a gold single crystal in which the longitudinal wave was propagated along a [110] crystal axis. The solid line on the graph has the slope predicted by the theory. It is obvious that the theory and experiment agree both qualitatively and quantitatively. Changing the field direction in the plane perpendicular to the propagation direction showed no anisotropy and simply reproduced the data shown.

The theory also predicts that the sound velocity change should depend on the square of the sine of the angle between the propagation direction and the magnetic field for a longitudinal wave and on the square of the cosine of the angle for shear waves. Figure 2 shows data on gold where the velocity change produced by the application of a 21-kOe field is plotted against the angle between the field and the propagation directions. The solid lines are plots of cosine squared and sine squared functions. Again the agreement is excellent.

For the cases discussed above, the inequality $\delta < \lambda$ was satisfied and the magnitude of the effect was independent of the material conductivity and determined solely by the value of ρC_0^2 in the particular specimen. This inequality is not satisfied in many cases for which the electrical conductivity is not very high or the sound waves have short wave length. The column labelled $1/8\pi\rho C_0^2$ in Table I gives the value of the coefficient of H^2 in the high-conductivity or long-wavelength limit while the column labeled γ_c gives the value of this coefficient calculated using values of the electrical conductivity given in the *American Institute of Physics Handbook*.[7] (The conductivity of the vanadium sample was measured in this laboratory using the method of Zimmerman.[8]) By comparing these two columns, the magnitude of the correction introduced by a finite conductivity or a short wavelength can be seen. The column labeled γ_e gives the experimentally measured values of the coefficient of the H^2 term as determined from the slope of the straight line on a $\Delta C/C$ vs H^2 plot. Comparison of γ_e and γ_c shows the degree of agreement between the macroscopic theory and the measurements. The last column presents the difference between theory and experiment and is to be compared with the experimental error in measuring the coefficient of H^2 of $\pm 0.1 \times 10^{-14}$ Oe^{-2}. In all the cases where $\delta \ll \lambda$, agreement is within experimental error. Agreement is fair in the cases where a correction for finite conductivity was used. This would indicate that the conductivity correction in the macroscopic theory is nearly correct but not quite.

[7] *American Institute of Physics Handbook* (McGraw-Hill Book Company, Inc., New York, 1957), p. 5–204.
[8] J. E. Zimmerman, Rev. Sci. Instr. **32**, 402 (1961).

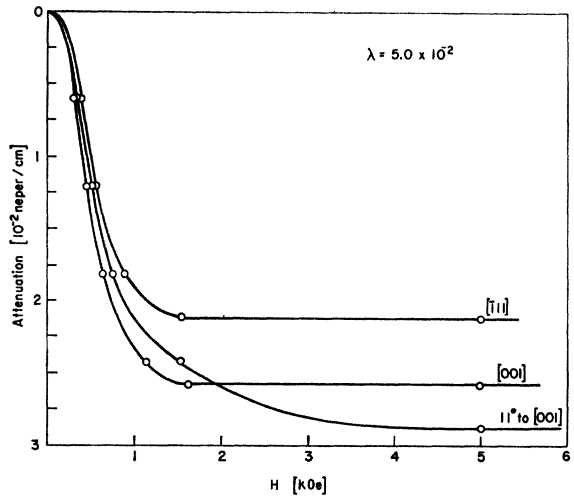

FIG. 3. Variation of the attenuation in amplitude of a 10-Mc/sec longitudinal wave propagating along a [110] crystal axis in a very pure copper crystal as a function of field strength. The field was always perpendicular to the propagation direction and directed along the crystallographic directions indicated.

The effect of the specimen temperature enters the theory mainly through the correction for finite conductivity so that lowering the temperature (raising conductivity) should make the magnitude of the effect approach a nearly temperature independent value of $1/8\pi\rho C_0^2$. Table II gives the results of measurements on longitudinal waves in the noble metals at 300, 77, and 4.2°K. In all these cases, the finite conductivity correction is negligible and the measured γ values are indeed temperature independent as predicted.

The macroscopic theory is based on the assumption that the electrons in the metal follow exactly the mechanical movement produced by the sound wave. This is certainly not the case when the electron mean free path becomes comparable to the sound wave length. It is, therefore, of vital interest to determine experimentally just how the sound velocity is changed by a magnetic field in a specimen in which this approximation is no longer valid. Such an experiment is complicated by the fact that under these same conditions the electrons absorb energy from the acoustic wave in a way that depends not only on the magnitude of an external field but also on its crystallographic direction. Figure 3 shows how the ultrasonic attenuation of a longitudinal wave was observed to vary with field in a special high purity copper crystal at 4.2°K. From the magnitude of the total attenuation change and the calculations of Steinberg,[9] it is possible to estimate that the ratio of electron mean free path to acoustic wavelength (at 10 Mc) is of the order of 0.08 in this sample. Unfortunately, these attenuation changes make the behavior of the "sing-around" system somewhat dubious because its oscillation frequency depends not only on the acoustic pulse transit time but also on the pulse height. This pulse height sensitivity can be compensated for by changing the gain in the oscillator loop but there is no assurance that such a gain change will not introduce a frequency shift by itself. It is, therefore, safest to operate the "sing-around" system under conditions of no attenuation change. For this reason, the field induced velocity changes were measured only at fields higher than 6 kOe and at a fixed orientation.

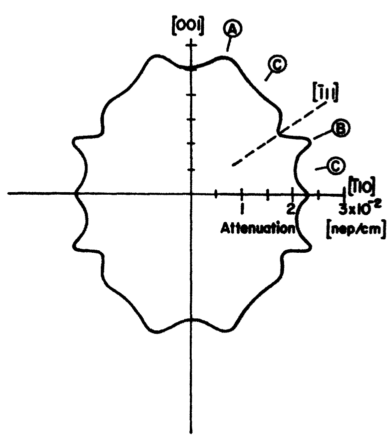

FIG. 4. Polar plot of the attenuation change produced by the application of a 17.5-kOe magnetic field to the pure copper crystal. The propagation direction of the longitudinal sound wave involved is along a [110] direction. The angles A, B, and C denote directions along which the magnetic field dependence of the velocity of sound was measured.

TABLE II. Temperature dependence of the coefficient of the H^2 term in Eq. (5) for the noble metals. For these metals $\delta \ll \lambda$. Units of 10^{-14} Oe^{-2}.

	Cu	Ag	Au
300°K	2.5	1.8	1.8
77°K	2.5	1.9	1.8
4°K	2.6	1.9	1.7

The choice of orientation is best discussed in terms of Fig. 4 which shows a polar diagram of the difference in attenuation between 0 and 17.5 kOe for various field directions in the plane perpendicular to the propagation direction. Figure 5 shows the change in the velocity of a longitudinal sound wave relative to its value at 6 kOe as a function of magnetic field at the various angles designated on Fig. 4. The solid curve is the parabola predicted by the macroscopic theory applicable at short electron mean free paths. It appears that in the principal crystallographic directions (except possibly the [110] direction) the macroscopic theory satisfactorily accounts for the observed variation of the sound velocity. If the field is along those directions for which the attenuation change is a maximum (directions A and B in Fig. 4), the velocity appears to vary linearly with applied field. Other nonprincipal directions (marked on Fig. 4 with the letter C) were also studied but these directions acted like the principal directions giving the same H^2 dependence predicted by the theory.

Figure 6(a) presents an enlarged polar plot of one

[9] M. S. Steinberg, Phys. Rev. 111, 425 (1958).

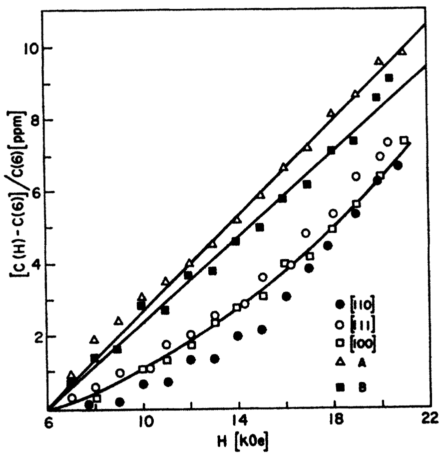

FIG. 5. Variation of the velocity of sound with magnetic field in the constant attenuation region above 6 kOe for various field directions in the plane perpendicular to the [110] propagation direction. Longitudinal waves in copper.

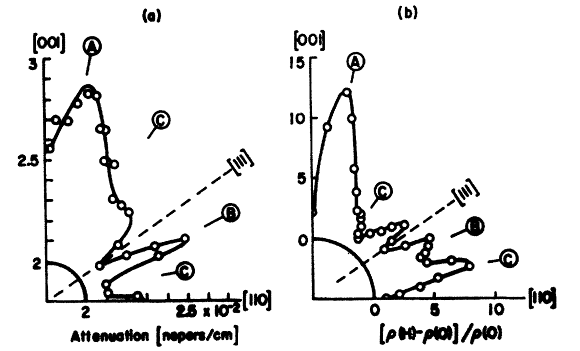

FIG. 6. (a) Magnified polar plot of the attenuation change accompanying the application of a 17.5-kOe magnetic field. (b) Variation of electrical resistivity with field direction in pure Cu at 23 kOe (data of reference 10).

quadrant of the attenuation versus magnetic field direction data shown in Fig. 4. The field directions along which the velocity of sound varies as the first power of the field are marked by the letters *A* and *B*. This plot is very similar to the polar diagram obtained by Alekseevskii and Gaidukov[10] who measured the variation of the electrical resistance of a high-purity copper crystal in which the current flowed along the [110] crystal axis and a field of 23 kOe was rotated in a plane perpendicular to the current direction. Figure 6(b) shows their results with the directions A and B superimposed. Alekseevskii and Gaidukov point out that in the directions in which $\Delta\rho/\rho$ is a maximum, the resistivity increases nearly with the square of the magnetic field while the directions of minimum $\Delta\rho/\rho$ are those in which the resistivity "saturates" by approaching a field independent value. The former directions are "open orbit" directions for the electrons.[11] It appears by comparing Figs. 6(a) and 6(b) that the velocity of sound varies linearly with magnetic field in two of the several "open orbit" directions. The reason that the sound velocity picks out only two directions is not understood. However, it is important to realize that in the magnetoresistance measurements the current was restricted to a [110] crystal direction while in the acoustic measurements it is only the sound wave propagation direction that is fixed parallel to [110]. Equation (1) indicates that in the acoustic experiment the current direction is at right angles to the propagation direction and the magnetic field so that as the magnetic field is rotated, the current changes crystallographic directions.

CONCLUSION

The data shows that as long as the electron mean free path and the electromagnetic penetration depth are short compared to the acoustic wavelength, the macroscopic theory of Alpher and Rubin provides a quantitative description of all the observations. In cases where the penetration depth is comparable to the wave length the theory predicts a correction factor which is very slightly smaller than needed for perfect agreement. For the case in which the mean free path is comparable to the wavelength, the macroscopic theory still seems to apply except in certain particular directions of the magnetic field. These directions correspond to at least some of the open orbit directions appearing in magnetoresistance measurements. In these special directions the velocity of sound varies linearly with magnetic field between 6 and 21 kOe.

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[10] N. E. Alekseevskii and Yu. P. Gaidukov, Soviet Phys.—JETP **10**, 481 (1960).

[11] R. G. Chambers, in *The Fermi Surface*, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960), p. 100.