Electron Relaxation Time Anisotropy in Copper*

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A method for determining electron relaxation time anisotropies in metals is suggested and presented along with preliminary data taken at 4.2°K using a single crystal grown from 99.999% pure copper. The analysis is based on the behavior of the attenuation of ultrasonic waves by conduction electrons in the high magnetic field limit. The data are discussed in terms of the Pippard model of the Fermi surface of copper. The technique also allows a rather direct test of the free electron theory of ultrasonic attenuation in that shear wave wave measurements are used in determining the total attenuation caused by the conduction electrons. It is suggested that a study of high-field shear wave attenuation will allow the total electronic attenuation to be found in any metal, whereas previously it has been possible to determine this quantity only for superconductors. On the rough experimental model used it is found that the relaxation time of electrons in neck orbits is several times larger than that of the other orbits studied. The relaxation times are of the order of 10^{-10} sec, and are impurity limited at 4.2°K. Mean free paths are found to be about 20 times smaller than estimated from the number of magnetoacoustic oscillations.

I. INTRODUCTION

VARIOUS properties of the conduction electrons in metals can be investigated by studying the attenuation of ultrasonic waves in single-crystal samples in the presence of an external magnetic field.[1] The period of oscillation of the attenuation in a magnetic field H at liquid helium temperatures can give information about the Fermi surface of the metal, a technique now known as the magnetoacoustic effect. Up until the present time the majority of investigations utilizing this technique has consisted of data on the Fermi momenta. The present work, however, illustrates another facet of the ultrasonic technique which in favorable cases could contribute to a thorough study of the electronic properties of a particular metal.

It is possible to obtain at least qualitative information about electron relaxation time anisotropy from an investigation of the variation of high-field attenuation as a function of frequency of the sound wave.[2] The analysis of the data is based on the free electron theory of metals but should give a good estimate of how electron relaxation times vary over the Fermi surface. Since very few data on relaxation times are presently available, possibly the present technique will lead to more detailed studies on some metals which have well-known Fermi surfaces. The present study is derived in conjunction with regular magnetoacoustic data and, therefore, a knowledge of the Fermi surface is important in the interpretation of the results. The relative simplicity of the Fermi surface of copper; in particular, the fact that it is contained in one band, makes copper an ideal metal for such an investigation. Application of the present technique also allows a direct test of the validity of the free electron theory of ultrasonic attenuation which gives an indication of how much confidence can be placed in this method. It is to be stressed, however, that the experimental data presented in this paper are preliminary and are given more for illustration of the suggested method than for the numbers obtained.

Information about relaxation time anisotropy could also be gained from cyclotron resonance or de Haas–van Alphen measurements. Of these two it appears that the cyclotron resonance technique is better suited for an exhaustive study of relaxation times. Unfortunately, the mass spread measured in cyclotron resonance experiments in copper cannot be separated readily from relaxation time anisotropy effects. It is possible that cyclotron resonance measurements at higher microwave frequencies than those presently used will allow these effects to be separated, so that relaxation time anisotropy could be studied.[3] Studies of the variation in amplitude of de Haas–van Alphen oscillations can, in principle, give information about relaxation times. This has not, however, proved possible because the magnetic field variation of amplitude is modified considerably by slight sample imperfections which cause complicated interference effects.[4]

II. THEORY

Complete theories of ultrasonic attenuation in metals at low temperatures have been given for the zero magnetic field free electron case,[5] for free electrons in the presence of a magnetic field,[6–8] and for the case of a real metal.[9] For a longitudinal sound wave propagating

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through a metal crystal in zero field it is found that the ultrasonic attenuation caused by conduction electrons on the free electron model is

$$\alpha_L(ql)=\frac{nm}{\rho v_s\tau}\left[\frac{q^2l^2\tan^{-1}ql}{3(ql-\tan^{-1}ql)}-1\right], \tag{1}$$

where n is the number of free electrons per unit volume, m is the electron mass, v_s is the sound velocity, ρ is the metal density, τ is the electron relaxation time, q is the magnitude of the sound wave vector ($q=2\pi\omega/v_s$), ω is the sound wave frequency, and l is the electron mean free path.

In the presence of a magnetic field H, the compressional attenuation based on a free electron derivation is found to be[7]

$$\alpha_L(H)=\frac{nmvq^2l}{3\rho v_s}\times\{[1-g_0(\beta k_0)+\tfrac{1}{4}g_0'^2(\beta k_0)/s_0(\beta k_0)]^{-1}-1\}, \tag{2}$$

where k_0 is the radius of the free electron Fermi sphere, $\beta=\hbar q/eH$, e is the electron charge, and $g_0(\beta k_0)$ $g_0'(\beta k_0)$, and $s_0(\beta k_0)$ are the functions introduced by Cohen, Harrison, and Harrison.[7] The appearance of Bessel functions in the expressions g_0, g_0', and s_0 leads to the oscillatory variation of the attenuation with magnetic field, i.e., the magnetoacoustic effect. The present analysis is not explicitly concerned with the oscillatory effect but rather with the behavior of the longitudinal attenuation $\alpha_L(H)$ as $H\rightarrow\infty$. In the free electron case this is given by[9]

$$\lim_{H\rightarrow\infty}\alpha_L(H)=nmvq^2l/15\rho v_s. \tag{3}$$

It is seen that the high-field limit of the attenuation is proportional to q^2l. A calculation of the type indicated by Pippard[9] shows that this limit is approached as H^{-2}.

Experimentally, it is possible to determine the difference between Eqs. (1) and (3) by comparing the attenuation at the high-field limit with that in zero field. When the high-field attenuation is equal to the attenuation in zero field, i.e., when

$$\frac{nmvq^2l}{15\rho v_s}=\frac{nm}{\rho v_s\tau}\left[\frac{q^2l^2\tan^{-1}ql}{3(ql-\tan^{-1}ql)}-1\right],$$

a numerical calculation shows that $ql=6.8$. Thus, if the frequency at which $\alpha_L(H\rightarrow\infty)=\alpha_L(H=0)$ can be found, the electron mean free path l can be determined. The relaxation time τ is related to l by $v\tau=l$, where v is the velocity of the electron on the Fermi surface.

In order to relate the present data to theory, the model of the Fermi surface of copper proposed by Pippard[10] on the basis of anomalous skin effect measurements will be used. This model has been generally substantiated by de Haas–van Alphen,[4] cyclotron resonance,[3] and magnetoacoustic[11] data as well as by theoretical calculations.[12,13] The model consists roughly of a sphere with eight protruding necks which touch the Brillouin zone boundary in the [111]-type directions. Because contact with the zone boundary occurs, the copper Fermi surface can be thought of in the repeated zone scheme as a body-centered array of spheres joined along [111]-type directions by the protruding necks.[11] It follows that any extremal orbit formed by planes cutting the Fermi surface perpendicular to H can give rise to magnetoacoustic data and can thus be studied by the present technique. Well-known orbits on this model include the "dog's bone," "neck," and "belly" orbits. The dog's bone orbit corresponds to a path around four adjoining cells by way of the necks and thus involves four zone boundary reflections. The belly orbit is that around the main body of the sphere, while neck orbits are those observed when H is along [111] and are orbits around the protruding necks. Another orbit included in this paper is a noncentral extremum running over two necks at the flat end of the dog's bone.[11] This orbit is produced along with a dog's bone rotated 90° when the sound propagation is along [110] and the magnetic field along [1$\bar{1}$0]. It is to be noted that data taken for neck orbits is a combination of more than one type of electron orbit since for **H** along [111] several extremal orbits on the Fermi surface of copper are possible. The same situation might also exist for the other orbits. The extremal orbits are primarily responsible for the attenuation[9] but others might well contribute.

III. EXPERIMENTAL PROCEDURE

The experiments were performed at the boiling point of liquid helium using a single crystal grown from 99.999% pure copper which was cut so that sound could be propagated along either the [001] or the [110] direction. The sample was prepared by James F. Kirn of the Virginia Institute for Scientific Research. Electron relaxation times in it are impurity limited at 4.2°K.

The ultrasonic apparatus used is similar to that of Morse[14] except that the present data were automatically recorded. A pair of 5- or 10-Mc/sec quartz transducers were bonded to the copper sample with Nonaq, one transducer serving to transmit the sound wave and the other to receive it. Sound wave frequencies from 30 to 100 Mc/sec were employed in the measurements. The magnetic field was supplied by a 7-in. Harvey-Wells electromagnet which produces magnetic field intensities

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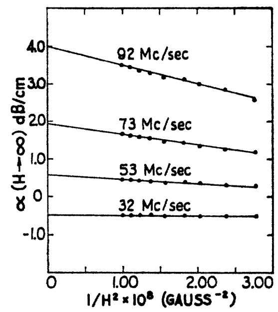

FIG. 1. High-field relative attenuation vs $1/H^2$ for different frequencies for compressional waves in copper. Sound propagation is along [110] and **H** is along [001] so that extremal orbits are belly orbits.

up to 11 000 G with the particular pole faces and gap used here.

Measurements for the present work were made in the following manner: The sound propagation direction and magnetic field direction were selected to correspond to the particular electron orbit to be studied. The relative attenuation of the ultrasound reaching the receiving transducer after passing through the sample was then recorded as a function of magnetic field up to the maximum field available. The attenuation values relative to the $H=0$ values were calibrated by a precise voltage measurement. The above procedure was then repeated for several sound wave frequencies for each electron orbit to be studied.

IV. RESULTS

Data were taken at a variety of frequencies for sound propagation along [001] and [110] directions. The following orbits were studied:

(A) Dog's bone: **q** along [001], **H** along [110].
(B) Belly: **q** along [001], **H** along [010].
(C) Belly[15]: **q** along [110], **H** along [001].
(D) Neck: **q** along [110], **H** along [111].
(E) Extremum over two necks: **q** along [110], **H** along [1$\bar{1}$0].

The first step in the analysis of the data is to determine the high-field limit of the attenuation for each of the orbit configurations above. Since the limiting attenuation at infinite field is approached as H^{-2}, high-field relative attenuation values were plotted against H^{-2} for each frequency used. One set of these data, that for belly orbits with **q** along [110] and **H** along [001], is shown in Fig. 1. It is seen that for this particular case the points fall closely on straight lines which can be extrapolated to infinite field. In some instances, however, it appeared that the high-field attenuation was still climbing rather steeply at 11 000 G, the highest field available with the present equipment. This naturally introduces uncertainty in the extrapolation to infinite field and it is seen then that the present technique will be effective only if the high-field limit of attenuation is reached. It was possible, however, with the present data to establish limits on the high-field values and this procedure was followed. Figure 2 is a plot of the extrapolated attenuation values as a function of sound wave frequency for three of the orbits studied. The approximate frequency at which $\alpha_L(H\to\infty)=\alpha_L(H=0)$ can be determined in each case. This frequency ω_s is just that for which $ql=2\pi\omega_s l/v_s=6.8$ and thus it is directly related to the electron mean free path for the orbit. The values of l calculated for the different orbits are given in Table I.

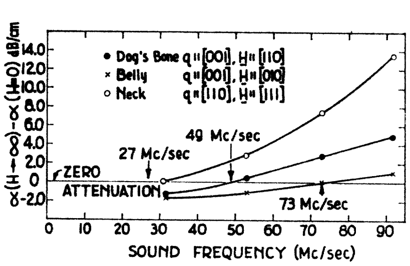

FIG. 2. Values of relative attenuation extrapolated to infinite field as a function of sound frequency for three of the orbits studied. The frequency ω_s at which $\alpha_L(H\to\infty)=\alpha_L(H=0)$ is indicated for each orbit.

It is possible to check the agreement of the above data with the free electron theory of the attenuation by making a plot such as Fig. 3. In Fig. 3 is plotted the normalized attenuation α_L/α' as a function of ql, where

$$\alpha'=\pi nmvq/6\rho v_s \tag{4}$$

is the zero-field attenuation for large ql. The curved line is just Eq. (1), the attenuation in zero field, and the straight line is Eq. (3), the attenuation as H approaches infinity. The quantity which is measured should be the difference between these two curves.

The present data can be fitted to Fig. 3 by normalizing the frequency at which each orbit reaches $ql=6.8$ to the intersection of the two curves. The attenuation values are normalized either by taking the best fit of the data, or by use of independent shear wave measurements as is done here.

It has been shown theoretically[9] that the attenuation of shear waves by the conduction electrons vanishes in certain cases as the magnetic field becomes very large, suggesting that the total electronic attenuation can be determined by shear wave measurements at high fields. The total attenuation caused by the electrons is, however, just α' of Eq. (4) if the condition of large ql is satisfied. An analysis of the high-field shear wave attenuation in copper has been carried out by Deaton[16] who found that the high-field attenuation approaches a value which is approximately the same for runs with various polarizations and field directions. It was assumed that ql was very large at 130 Mc/sec and α'

[15] This orbit was called by Morse (reference 11) a noncentral diamond-shaped orbit but is interpreted in the present work as a belly orbit since the dominant momentum value of the orbit corresponds closely to that expected for the body of the copper Fermi surface.

[16] B. C. Deaton, doctoral dissertation, The University of Texas, Austin, Texas, 1962 (unpublished).

TABLE I. Quantities pertinent to the relaxation time calculations along with results.

Orbit	ω_s (Mc/sec)	l (cm)	p from magneto-acoustic data (g cm/sec$\times 10^{19}$)	τ using $v_0 = 1.58\times 10^8$ (sec)	Segall's theoretical values of m^* (units of m_e)	τ from theoretical m^* values (sec)	m^* from cyclotron resonance (units of m_e)	τ using cyclotron resonance m^* (sec)
Dog's bone $\mathbf{q}\parallel[001]$, $\mathbf{H}\parallel[110]$	49	0.0097	1.30	6.2×10^{-11}	1.12	7.6×10^{-11}	1.22	8.3×10^{-11}
Belly $\mathbf{q}\parallel[001]$, $\mathbf{H}\parallel[100]$	73	0.0065	1.40	4.1×10^{-11}	1.1[a]	4.5×10^{-11}	1.26[b]	5.3×10^{-11}
Belly $\mathbf{q}\parallel[110]$, $\mathbf{H}\parallel[001]$	47	0.012	1.42	7.6×10^{-11}	1.1[a]	8.5×10^{-11}	1.39	$10\ \times 10^{-11}$
Neck $\mathbf{q}\parallel[110]$, $\mathbf{H}\parallel[111]$	27	0.020	0.28	$13\ \times 10^{-11}$	0.41	$27\ \times 10^{-11}$	0.6	$40\ \times 10^{-11}$
Extremum over two necks $\mathbf{q}\parallel[110]$, $\mathbf{H}\parallel[1\bar{1}0]$	42	0.013	1.06	8.2×10^{-11}				

[a] Segall assumed a constant m over the belly.
[b] This effective mass is measured for a slightly different part of the belly than that referred to in the present work.

found to be approximately 6 dB/cm at that frequency. The results shown in Fig. 3 are normalized on this basis and it is seen that a fairly good fit of the data to the free electron model is obtained.

In addition to providing the normalization above, it should be emphasized that the shear measurements offer the possibility of a straightforward method for determining the total electronic attenuation in any metal sample used. One reason that so few data on the absolute magnitude of the electronic contribution to ultrasonic attenuation have been published is that one must determine how much of the attenuation is caused by nonelectronic absorption mechanisms. Relative measurements of attenuation are always much easier to carry out, so the technique of "wiping out" the electronic attenuation with large magnetic fields should be useful for normal metals. The only other means of measuring the total electronic attenuation can be applied only to superconductors and relies on the fact that the electronic attenuation in the superconducting state approaches zero as the absolute temperature approaches zero.[17]

The calculation of electron relaxation times for the various orbits in copper will be carried out in two ways. The first method involves the assumption that the Fermi velocity v is constant over the entire Fermi surface of copper. This admittedly is a crude approximation, especially for orbits which involve zone boundary reflections or pass over parts of the Fermi surface which are near the Brillouin zone boundary. The second method involves calculating the relaxation time using effective mass data from cyclotron resonance measurements[3] and theoretical calculations by Segall.[12]

Assuming an average electron velocity over the Fermi surface is equivalent to saying that the relaxation time anisotropy is just that of the mean free path since these are related by $v\tau = l$. Since the present data are being fitted to a free electron theory, the average electron velocity used in the calculation is that calculated from the radius of the free electron sphere in copper. The free electron sphere momentum $p_0 = mv_0 = 1.44 \times 10^{-19}$ g cm/sec, thus $v_0 = 1.58\times 10^8$ cm/sec. The values of τ calculated in this fashion are given in Table I for the five orbits investigated. It is seen that the relaxation times calculated are of the order of 10^{-10} sec. The

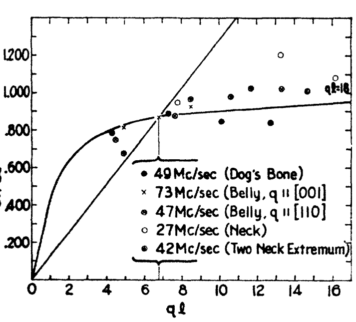

FIG. 3. Normalized relative attenuation of compressional waves in copper plotted vs ql for the different orbits studied. The straight line represents $\alpha_L(H\rightarrow\infty)/\alpha'$, and the curve represents $\alpha_L \times (H=0)/\alpha'$ for the free electron model. The experimental values of $[\alpha_L(H\rightarrow\infty)-\alpha_L(H=0)]/\alpha'$ are plotted from the straight line. If the free-electron treatment is correct, they should fall on the curves. The frequencies ω_s are normalized to $ql=6.8$.

[17] R. W. Morse and H. V. Bohm, Phys. Rev. **108**, 1094 (1957).

neck relaxation time is roughly twice as large as relaxation times for the other orbits. The shortest relaxation time on this assumption is for the belly orbit with **q** along [001] and **H** along [010].

It is possible to calculate the relaxation time τ in a different manner. With our rough model we introduce the concept of effective mass m^* into the free electron equations to obtain

$$\tau = m^* l / p, \tag{5}$$

where p is the Fermi momentum associated with the orbit under consideration. The values of the Fermi momenta perpendicular to both **q** and **H** for the five orbits were measured by ordinary magnetoacoustic techniques[16] and are given in Table I. The values of effective mass m associated with the orbits under consideration are taken from experimental cyclotron resonance data[3] and from theoretical values calculated by Segall[12] who studied the energy bands in copper by the Green's function method. There is a disagreement between the experimental and theoretical values of effective mass of about 30%. This discrepancy is thought to be caused by the omission of certain real metal effects from the theory.

The values of τ calculated from Eq. (5) and the values of m^* used are given in Table I. The agreement with the previous calculation is seen to be reasonable, the only large discrepancy being for the neck relaxation time, a disagreement which is not unexpected since there is a fairly large uncertainty in the m^* values and since this orbit occurs in a region where a large departure from the free electron model should occur. It is seen from Table I for this calculation the neck relaxation time is also much larger than the relaxation time in other orbits. This calculation thus appears to give a result contradictory to the rough theoretical prediction of Ziman[18] who suggests that τ_{neck} is less than τ_{belly} in copper. Ziman's calculation was prompted by the disagreement between theoretical and experimental values of Hall effect, magnetoresistance, and other measurements in copper.[19]

V. CONCLUSIONS

It is seen that the present technique allows a direct test of the validity of the free electron ultrasonic attenuation calculations, and also can provide the most direct experimental information presently available on electron relaxation time anisotropy in a metal. It appears that if the Fermi surface of a metal is simple and relatively well known, it will be possible to study τ with this technique. The present calculations are rough, of course, but since there are no data at all available on relaxation time anisotropy, possibly even this rough an estimate is worthy of note.

The use of shear wave measurements is suggested as a possible means of determining the amount of acoustic attenuation caused by the conduction electrons. This would allow the total electronic attenuation to be found in any metal, whereas before it has been possible to determine this quantity only for superconducting metals.

Although the experimental data included are considered preliminary and given primarily as an illustration of the suggested technique, certain conclusions can be drawn. The relaxation time of electrons in neck orbits is several times larger than that of any other orbit investigated. The shortest relaxation time measured is that of electrons in belly orbits. This is confirmed both for the simple assumption of a constant Fermi velocity over the Fermi surface and calculations based on both experimental and theoretical effective mass values for the orbits under consideration.

A rather direct calculation of the electron mean free path is obtained for the orbits analyzed. The values of l are found to be approximately twenty times smaller than those found by calculations based on the number of magnetoacoustic oscillations obtained, or the lowest magnetic field value for which an oscillation occurs. It, thus, appears that the method of calculating l used by Galkin and Korolyuk[20] as well as others must be replaced by a more careful analysis. This is not surprising since the field value at which oscillations cease to be observable depends also upon the sensitivity and noise level of the experimental apparatus. "Mean free path" implies a certain exponentially decreasing probability of scattering, as is quite apparent in the curves calculated by Kjeldaas and Holstein[6] for finite ql values. Even for $ql=7$, or wavelength approximately equal to mean free path, at least two oscillations are seen, whereas under the simple interpretation one would expect none.

It would be of interest to make a similar study in a sample where the relaxation times are phonon limited. One could then measure variations in relaxation times as a function of temperature.

ACKNOWLEDGMENTS

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[18] J. M. Ziman, Phys. Rev. **121**, 1320 (1961). *Note added in proof.* Dr. Ziman has kindly pointed out to us that there is no disagreement if the impurities are homovalent with copper.

[19] J. M. Ziman, in *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1961), Vol. **10**, p. 1.

[20] A. A. Galkin and A. P. Korolyuk, J. Exptl. Theoret. Phys. (U.S.S.R.) **38**, 1688 (1960) [translation: Soviet Phys.—JETP **11**, 1218 (1960)].