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Determination of Relaxation Times by Magnetoacoustic Measurements in Copper†

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Experimental and theoretical evidence is presented to support the hypothesis that the amplitudes of observable magnetoacoustic oscillations are to a large extent controlled by collision damping. By fitting experimental data on copper to a simple formula derived from the free-electron theory, we find that the temperature dependence of the electron-scattering rate for belly orbits is given by $(1.5 \pm 0.2)\times 10^6 T^3$ for temperatures below about 13 °K. This value is comparable to those obtained by other workers using cyclotron resonance. Although our method needs further theoretical development, we believe it has considerable potential as a tool for investigating scattering processes in metals.

I. INTRODUCTION

In a recent paper, Häussler and Welles[1] demonstrated that numerical values for the electron relaxation time in metals can be extracted from measurements on the amplitudes of Azbel'-Kaner cyclotron-resonance (AKCR) oscillations. The amplitude A_n of the nth resonance peak, is, according to free-electron theory, proportional to $n^2 e^{-2\pi n/\omega\tau}$, where ω is the microwave frequency and τ is the relaxation time. Since the cyclotron frequency ω_c is given by $\omega = n\omega_c$, this behavior has the simple physical interpretation that the amplitudes are primarily determined by the probability $e^{-2\pi/\omega_c\tau}$ that an electron can complete an orbit and return to the skin region where the microwave field is large. (This simple behavior is only followed when one can neglect the possibility that an electron completes more than one orbit, i.e., when $e^{-2\pi/\omega_c\tau} \ll 1$.) Häussler and Welles were able to obtain values for the effective scattering rate in copper as a function of temperature for several distinct orbits on the Fermi surface.

In this paper, we demonstrate that the magnetoacoustic analog of Häussler and Welles's experiment can be carried out. The magnetoacoustic effect involves spatial rather than temporal resonances, in fact resonance peaks occur whenever some suitable dimension on an electron orbit is a multiple of the sound wavelength. Several workers[2–4] have reported an exponential decrease of the amplitude of the observed oscillations with harmonic index n and we conclude that the regime $e^{-2\pi/\omega_c\tau} \ll 1$ is experimentally accessible.

II. THEORY

A. Free-Electron Model

Cohen, Harrison, and Harrison[5] have laid down the theoretical groundwork for a wide range of magnetoacoustic effects using the free-electron model and assuming an isotropic relaxation time τ (i.e., the electron mean free path is given by $l = v_F\tau$, where v_F is the Fermi velocity). They showed that for longitudinal sound waves of wave vector $\vec{q}$ in the presence of a transverse magnetic field $\vec{H}$ (normal to $\vec{q}$) the attenuation coefficient is given by

$$\alpha = \frac{Nm}{\rho v_s \tau} S_{11}(X, ql) , \tag{1}$$

where N is the electron density; m is the electron mass; ρ is the density of the metal; v_s is the sound velocity; and the argument X of the function S_{11} is given by $X = qR$, where R is the orbit radius, in real space, of an electron on an equatorial belt of the spherical Fermi surface. In terms of the Fermi radius k_F, this orbit dimension is given by $R = c\hbar k_F/eH$, so that X varies as $1/H$. In the original paper the function S_{11} was computed by an approximate numerical method and it was shown to exhibit an oscillatory dependence on $1/H$ similar to that observed experimentally in real metals.

More recently, Gavenda and Chang[6] obtained an analytical expression for S_{11} in the limits $X \gg 1$ and $ql \gg 1$. The result is

$$S_{11} = \frac{\pi ql}{6}\left(\frac{\sinh(\pi X/ql)\left[\cosh(\pi X/ql) + (\pi X)^{-1/2}\cos(\pi\omega/\omega_c)\sin(2X - \frac{1}{4}\pi)\right]}{\sinh^2(\pi X/ql) + \sin^2(\pi\omega/\omega_c)}\right), \qquad (2)$$

where ω is the sound frequency, and is a good approximation over a wide range of values of X and ql provided that these are both fairly large.[7] It is to be noted that this expression predicts a null and a phase reversal when $\omega = \omega_c$ and that this effect was observed by Gavenda and Chang in cadmium. In the present context, however, it is of greater interest to evaluate Eq. (2) subject to the further limitations $\omega \ll \omega_c$ and $e^{-2\pi X/ql} \ll 1$. The result is

$$S_{11} = \tfrac{1}{6}\,\pi ql\left[1 + 2(\pi X)^{-1/2} e^{-\pi X/ql}\sin(2X - \tfrac{1}{4}\pi)\right] . \qquad (3)$$

This simple expression indicates that the attenuation consists of a constant part, $\frac{1}{6}\pi ql$, together with magnetoacoustic oscillations in the form of a damped sinusoid. The damping is characterized by a geometric damping factor, proportional to $X^{-1/2}$, and a collision damping factor $e^{-\pi X/ql}$.

The advantage of expressing the predictions of the free-electron model in this particular form becomes apparent when we consider the physical interpretation, which is very clear.

(a) The magnetoacoustic oscillations, in this low-field limit, complete a cycle whenever $2R = n\lambda$, i.e., the periodicity corresponds to the extremal caliper dimension on the Fermi surface, as expected. Thus the oscillations are associated with a belt of electrons on the equatorial region of the spherical Fermi surface. The thickness of this belt (measured in a direction parallel to $\vec{H}$) is governed by the fact that orbits of less than extremal diameter produce oscillations which are not in phase with those from the dominant extremal orbits, and it is straightforward to show that for the spherical geometry this criterion leads to a belt thickness proportional to $X^{-1/2}$.

(b) The relaxation-time assumption commonly used in transport theory is equivalent to the statement that an electron has a probability $e^{-t/\tau}$ of surviving for a time t without a collision. The collision damping factor, which can be expressed in the equivalent form $e^{-\pi/\omega_c\tau}$, can therefore be interpreted as the probability that an electron completes exactly half an orbit, which is in fact the minimum required to span the distance between caliper points. This is, of course, to be expected, because the electron interacts strongly with the sound wave every half-orbit, where it runs parallel to the planes of constant acoustic phase.

B. Real Metals

The problem of the magnetoacoustic effect in real metals has been treated at length by Pippard[8] but the case $e^{-2\pi/\omega_c\tau} \ll 1$ was not discussed explicitly. In the absence of a formal theoretical treatment, we will use the physical arguments outlined above to predict the form of the magnetoacoustic oscillations in this limit. It is already well known[9] that in real metals the oscillations tend to be dominated by orbits on the Fermi surface whose extension in the $\vec{q}\times\vec{H}$ direction is stationary with respect to displacements normal to the plane of the orbit. Furthermore, it is a matter of common observation that the oscillations resemble a damped sinusoid at sufficiently small fields. We will, in addition, assume that:

(a) The damping can be characterized by a geometric damping factor and a collision damping factor as in the free-electron model. In the event that several periods are present, arising from different portions of the Fermi surface, the oscillations will combine additively and hence can, in principle, be separated by harmonic analysis.

(b) If the caliper points on the Fermi surface are simple limiting points characterized by principal radii of curvature, the geometric damping factor will be of the form $X^{-1/2}$ as in the free-electron model. Other cases, however, must be scrutinized individually on the basis of the phase cancellation process.

(c) The collision-damping factor must be applied in its most general form as the probability that an electron spans the distance between caliper points, where it interacts strongly with the sound wave. If the caliper points lie at the opposite ends of a simple closed orbit, the formulation $e^{-\pi/\omega_c\tau}$ is still appropriate, where the cyclotron frequency $\omega_c = eH/m^*c$ is expressed in terms of the cyclotron mass for the same orbit. These masses are well tabulated for many metals as a result of the extensive cyclotron-resonance measurements which have been made in the past. Since the caliper dimension on the Fermi surface is given by $\Delta k = en\lambda H_n/c\hbar$ it is possible to express the collision damping factor in a form appropriate to a magnetoacoustic measurement as $e^{-n\pi m^* \lambda/\tau\hbar\Delta k}$. This is the factor which modulates the amplitude A_n of the nth oscillation (in addition to the geometric damping factor $n^{-1/2}$). It is quite common[9] for magnetoacoustic oscillations to be associated with a dimension Δk on an open orbit running normal to $\vec{q}$ in real space, or with a nonextremal dimension Δk on a reentrant closed orbit. In such cases, the corresponding effective mass may not have been tabulated, but nevertheless can be defined in terms of the time taken by the electron in spanning the distance between caliper points.

It would therefore appear that numerical values for the relaxation time could be obtained by fitting experimentally determined oscillation amplitudes to our theoretical model. However, we must also remember that, in real metals, the scattering process may not be well represented by an isotropic relaxation time τ. Nevertheless, any experimentally determined value of this parameter can still be given the appropriate physical significance. For example, if the scattering can be described by a $\vec{k}$-dependent relaxation time, the measured $1/\tau$ value would represent an average of $1/\tau(\vec{k})$ around an orbit. On the other hand, for small-angle scattering such as that caused by low-temperature thermal phonons, the experimentally determined $1/\tau$ value would be a measure of the average *effective* scattering rate around the orbit. The question of the effectiveness of small-angle scattering in this particular transport process is quite complicated, and we will return to it later in the discussion of the experimental results.

III. EXPERIMENT

The ultrasonic attenuation measurements were made using the conventional pulse-echo technique[9] with the addition of a temperature control and measurement facility. The details of the method were very similar to those described in a recent paper by Witt and Peverley[10] in which the magnetoacoustic effect was observed as a function of temperature in potassium. In copper, however, the Debye temperature is much higher than in potassium, and so it was necessary to extend the upper limit of the temperature range to about 20 °K. Above this value, the magnetoacoustic oscillations were completely washed out by electron-phonon scattering. In the present work, longitudinal sound waves of frequency 165 MHz were propagated in a single crystal of copper having a resistivity ratio of about 44 000. The sound propagation vector $\vec{q}$ was along the [110] direction and the magnetic field $\vec{H}$ was rotated in the plane normal to $\vec{q}$ in order to select specified orbits on the Fermi surface. Very strong oscillations due to belly orbits were observed for $\vec{H} \parallel [001]$ and these oscillations formed

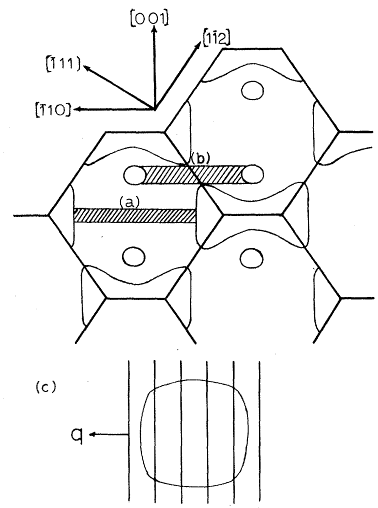

FIG. 1. (110) projection of the Fermi surface of copper with $\vec{H} \parallel [001]$ showing (a) belly orbits, (b) rosette orbits, (c) belly-orbit shape in relation to sound wave.

the principal object of study in the current investigation. It will be seen from the Fermi-surface projection in Fig. 1 that for this field direction, the four-cornered rosette orbit also exists, however, it turns out that the oscillations due to this orbit are so strongly damped that they interfere only with the first few belly-orbit oscillations. The latter could therefore be observed clearly for orders 5–26, beyond which the oscillation amplitudes were comparable to the noise level. We were thus able to make meaningful amplitude measurements on a sequence of about 20 consecutive oscillations, although this number diminished at higher temperatures. A tracing of some selected raw data is shown in Fig. 2. It can be seen that the oscillations become more nearly sinusoidal as n increases and that the damping increases with temperature.

We define the amplitude A_n to be the difference between the attenuation at the nth maximum and the average between two neighboring minima. A plot of $n^{1/2}A_n$ vs n is given in Fig. 3. The dependence is clearly exponential within experimental error, and the exponent increases with temperature. It may be verified from these data that the conditions under which Eq. (3) were derived are well satisfied for all points in the diagram.

We conclude that it is possible to extract relaxation times from the data. To do this we express the collision damping factor in the form $e^{-n\pi m^* \lambda/\tau\hbar\Delta k}$ and substitute numerical values for known quantities. We quote[11] the value $m^*/m = 1.37$ for belly orbits. Values for Δk, the [1$\bar{1}$0] belly dimension, and the [110] sound velocity (needed to calculate λ) were taken from Ref. 4. By Matthiessen's rule, the scattering rate $1/\tau$ due to phonons can be separated from that due to other mechanisms by

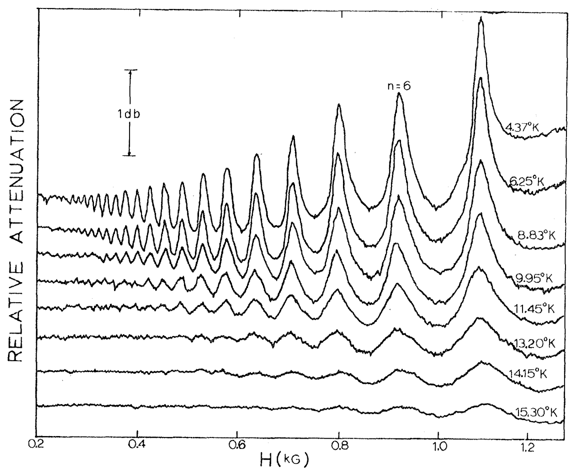

FIG. 2. Magnetoacoustic oscillations due to copper belly orbits at various temperatures. The increased rate of damping at higher temperatures is due to electron-phonon scattering.

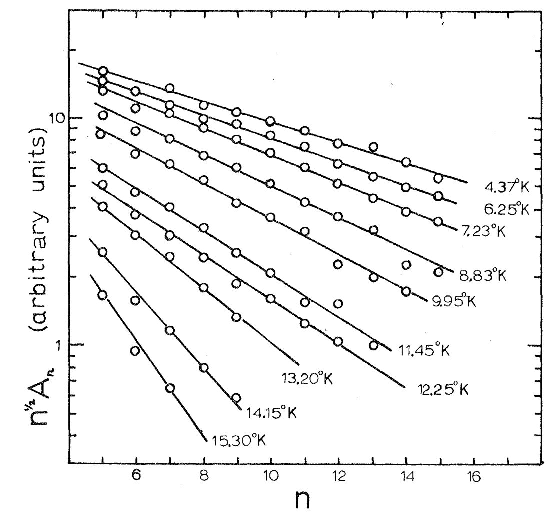

FIG. 3. Temperature dependence of the damping of the magnetoacoustic oscillation amplitudes for copper belly orbits.

simple subtraction. Since the phonon contribution is negligible below about 5 °K we subtracted all $1/\tau$ values from the value at 4.37 °K. The result is shown in Fig. 4. The phonon scattering rate exhibits a well-defined T^3 behavior, except at the higher temperatures. In numerical terms, we find that $1/\tau(T) - 1/\tau(0) = (1.5 \pm 0.2) \times 10^6 T^3$. This compares with the value $(2.9 \pm 1.2) \times 10^6 T^3$ measured by Häussler and Welles from their AKCR data and with the theoretical value $1.9 \times 10^6 T^3$ quoted by them.

In our sample, the residual scattering rate $1/\tau(0)$ was found to be 2.46×10^9. All scattering rates are expressed in reciprocal seconds.

IV. DISCUSSION

The preceding analysis completes our demonstration that the magnetoacoustic effect is capable of yielding essentially the same result as the AKCR experiment. Several aspects of this work merit further discussion.

A. Magnetoacoustic Oscillation Amplitudes

It is hoped that the evidence presented in this paper will clear up some of the confusion which has surrounded the subject of the magnetoacoustic oscillation amplitudes. The concept of the collision-damping factor was due originally to Pippard[12] but did not appear explicitly in his theory for real metals[8] where it was assumed that the probability of orbit completion must necessarily be large. Under these conditions, the damping is governed by geometric decay, which is relatively slow. Subsequently, several workers[2–4] reported that in their experimental data the decay was better described as exponential, and they obtained relaxation-time estimates by fitting to an assumed form for the collision damping factor. The numerical values they obtained may have been somewhat in error for two reasons: first, the assumption that an electron must remain unscattered for a whole orbit rather than half an orbit; and second, the omission of the geometric damping factor. However, their work did show that magnetoacoustic oscillations can be observed when the probability of orbit completion is small – a result which is endorsed in the present investigation.

More recently, Dooley and Tepley[13] obtained a numerical solution for the free-electron model and found that the oscillation amplitudes decayed exponentially. Deviations from true exponential behavior for small values of the harmonic index n were associated with the breakdown of the condition $\omega_c\tau < 1$. Similar behavior was observed in experi-

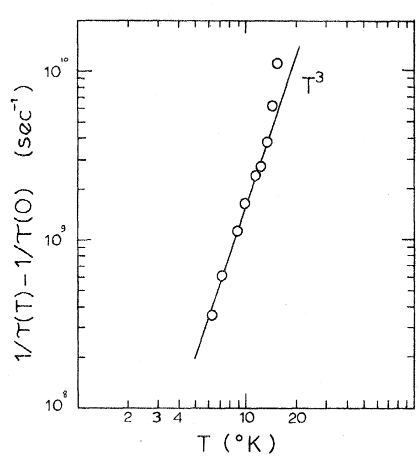

FIG. 4. Temperature dependence of the electron-scattering rate for copper belly orbits.

mental data on bismuth and they used this method to estimate the relaxation time in this metal. According to our analysis, deviations from Eq. (3) should be observed when the condition $e^{-2\pi/\omega_c\tau} \ll 1$ fails. Since this condition is several times less stringent than the condition $\omega_c\tau < 1$, we think it more likely that the deviations observed by Dooley and Tepley were due to the presence of the geometric decay factor, which they did not take into account.

B. Effectiveness of Small-Angle Scattering

The free-electron theory of Cohen, Harrison, and Harrison[5] was derived under the assumption of an isotropic relaxation time. In this model of the scattering process it is assumed that the position of an electron on the Fermi surface is randomized after a collision. When the scattering is due to low-temperature phonons, however, this is a very poor approximation to the truth. In fact, the change in wave vector suffered by an electron in a collision with a phonon is given roughly by $\delta k/k_F \sim T/\Theta$, where Θ is the Debye temperature, about 350 °K for copper. It is thus evident that in the current experiments the typical scattering angle is quite small. We thus need to consider seriously the question of exactly what is being measured in the present work.

In AKCR, the skin depth which characterizes the penetration of rf fields is of the order of 10^{-5} cm. This is about two orders of magnitude smaller than the orbit size for the fundamental AKCR, and proportionately more so for the higher harmonics. Thus, it is not difficult to see that even a small-angle phonon collision will prevent an electron from returning to the skin region, and will be just as effective as a collision which completely randomizes the electron on the Fermi surface. The scattering rate measured in the AKCR experiment is thus proportional to the total phonon density, which varies as T^3.

The case of the magnetoacoustic effect is not quite so straightforward, however, because the region of strong interaction with the sound wave is of the order of half an acoustic wavelength, about 10^{-3} cm in our case. This is of the same order as the orbit size for the fundamental geometric resonance, so that for small values of the harmonic index n a phonon would not perturb an electron orbit much in relation to the sound-wave pattern. Phonon scattering, in this limit, would not be very effective. On the other hand, for large n the region of strong interaction would occupy a small fraction of an orbit, and in the limit phonon scattering would become completely effective, as in AKCR.

The T^3 dependence of the electron scattering rate observed in our work suggests that phonon scattering is, for the most part, effective. However, the fact that our scattering rate is numerically less than that reported by Häussler and Welles (using AKCR) leaves some doubt as to whether we are, in fact, measuring the *total* rate. This question can, perhaps, be resolved by carrying out experiments at higher acoustic frequencies.

Some recent work by Witt and Peverley[10] is probably a good example of a magnetoacoustic measurement where phonon scattering is relatively ineffective. In this work, the temperature dependence of the scattering rate was determined by fitting experimental data on potassium (for which the Fermi surface is nearly spherical), to the free-electron model. Out of necessity, the measurements were restricted to small values of the harmonic index n, and it was found that, in order to obtain a good fit, it was necessary to postulate a mean free path which increased with magnetic field. This can be taken to mean, of course, that the effectiveness of phonon scattering increases with the harmonic index n, in agreement with our previous argument. In addition, it was found that the scattering rate increased as T^5. This implies that, in addition to the expected T^3 dependence due to the phonon density, there is a further dependence on the scattering angle (which increases as T) and this is qualitatively consistent with the conclusion that phonon scattering is not completely effective. (If it were, the measured scattering rate would not depend on the scattering angle.)

Our conclusion is, therefore, that the magnetoacoustic effect presents the interesting case of a transport process in which the effectiveness of small-angle scattering is a function of magnetic field. At small fields, where phonon scattering is fully effective, the total scattering rate can be measured by this means.

C. Anisotropy of Scattering Rate in Copper

There is a growing body of theoretical[14] and experimental[15] evidence that the phonon scattering rate in copper is highly anisotropic, with an exceptionally high rate on the necks. We attempted, as did Häussler and Welles,[1] to determine the neck rate by an examination of the neck orbits which occur when $\vec{H} \parallel [\bar{1}11]$. Unfortunately, the neck oscillations turned out to be so weak and highly damped that meaningful measurements could not be made without further improvements in the signal-to-noise ratio. From a qualitative examination of the neck oscillations, however, we could roughly estimate that the neck scattering rate is three or four times higher than the belly rate at all temperatures. This indicates a somewhat smaller degree of anisotropy than that observed by other workers, and a possible explanation may be that we were looking at orbits slightly displaced from the central plane of the neck, where the scattering rate is already reduced appreciably from its maximum value. The high scattering rate on the necks may also account for the heavy damping of the rosette-orbit oscillations, since this orbit is known to cross four necks.

V. CONCLUSIONS

The magnetoacoustic effect is a bulk measurement in a metal sample, so that surface effects need not be considered either in performing the experiment or in the interpretation of the results. In our relaxation-time studies, this is an advantage by comparison with methods based on cyclotron resonance, where the interaction occurs at the surface of the metal. On the other hand, small-angle scattering is not as effective in the magnetoacoustic effect as in AKCR, so it is necessary to restrict observations to high values of the harmonic index n in order to unambiguously determine the total electron phonon scattering rate. It is not clear that this limiting case has been achieved in the present work.

In spite of the aforementioned uncertainties, we believe that the magnetoacoustic effect has the potential for becoming a precision tool in studies of the electron scattering process. This potential will be realized more fully if some of the intuitive arguments we have been forced to rely upon can be justified by formal analysis.

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Scattering Effects in Photoelectric Emission from Solids. I*

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A simple model of photoelectric emission is solved essentially exactly. The model consists of a one-dimensional solid whose free-electron-like conduction band is cut off by a potential step representing the surface. Inside are sparse random local elastic scattering centers. The rate equations of Kane are derived rigorously, and hence expressions are obtained for the number of photoelectrons which escape with and without scattering as a function of the mean free path, the electromagnetic penetration depth, and the transmission coefficient of the surface barrier.

I. INTRODUCTION

The photoelectric effect is a powerful experimental tool for probing the electronic states of solids. Hopefully one can use it to gain information about the electronic density states of the bulk material. Two effects interfere with this aim: (a) Photoelectrons may originate near the surface where their initial wave function differs substantially from its bulk value; and (b) photoelectrons may scatter a number of times before emerging from the surface. The first effect can arise from the distortion of