mined calorimetrically assuming that $\gamma = 3.104$ mJ/mole K^2, is shown as a function of T. This shows that Θ decreases from its low-temperature value for temperatures above 9 K. The tendency toward slightly higher Θ values below 3 K than above this temperature is felt to be spurious and to result from the extreme sensitivity of the calorimetric value of Θ to small errors in the heat-capacity measurement when the lattice contribution is as small, as it is below 3 K. Although departures of Θ from the limiting value take place at slightly lower values of T/Θ than is typical of many metals, cases where deviations are found at even lower values of T/Θ are known to occur.[9]

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Electron Mean Free Path in Potassium*

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The magnetoacoustic effect was measured in potassium at temperatures of 1.2–8.5 °K. A quantitative comparison with the free-electron theory indicates that the mean free path at higher magnetic fields is effectively 1.1 to 1.25 times larger than at low fields. It is suggested that this deviation is due to the failure of the relaxation-time approximation to give an adequate description of the electron scattering. The temperature dependence of the mean free path was found to be in agreement with resistivity measurements.

INTRODUCTION

The purpose of this paper is to report measurements of the magnetoacoustic effect in potassium at temperatures ranging from 1.2 to 8.5 °K (above which the effect disappears). A quantitative comparison of the data with the free-electron theory of Cohen, Harrison, and Harrison[1] (CHH) is made. This comparison is used to determine the electronic mean free path (MFP) and its temperature dependence. A consistent deviation of the theoretical attenuation from the data is observed at higher magnetic fields at all temperatures. A qualitative argument indicates that this deviation could be due to the incorrectness of the relaxation-time assumption in the CHH theory.

EXPERIMENTAL PROCEDURE

The potassium samples were cylindrical disks $\frac{7}{16}$ in. in diameter and approximately $\frac{1}{4}$ in. long. These were spark cut from single-crystal boules grown from zone-refined stock by a method described by Foster *et al.*[2] The residual resistance ratios of the samples varied from 6000 to 8500. A single coaxially plated x-cut quartz transducer was bonded to each sample with Dow Corning 7 compound. Longitudinal waves were propagated along or near the [110]-growth direction. The samples were mounted in such a way that the propagation vector $\vec{q}$ was always perpendicular to the magnetic field $\vec{H}$, while the latter could be rotated through 360°. A pulse generator-receiver system was operated at 45 MHz to produce and detect the ultrasonic signals. A Matec model 1235 attenuation recorder was used so that the relative attenuation could be plotted directly as a function of magnetic field.

To obtain temperatures above the boiling point of liquid helium, heat was supplied by a resistance coil wrapped around the copper sample holder. The sample holder was insulated from the helium bath by a double-walled stainless-steel jacket, leaving a small residual coupling via the stainless-steel support tube. This coupling was necessary to maintain thermal equilibrium for a given heat input. Temperatures were measured with a germanium

resistance thermometer to within about 0.01 °K.

Sound velocities were determined from measurements of the sample lengths and the times between successive acoustic echoes as displayed on the oscilloscope.

COMPARISON WITH THEORY

Plots of the relative attenuation versus magnetic field were compared with the CHH theory of the magnetoacoustic effect. Assuming one free electron per atom, the attenuation coefficient is

$$\alpha = (Nmv_F/Mv_S l) S_{11}(X, ql), \quad (1)$$

where N is Avogadro's number, m is the electron mass, v_F is the Fermi velocity, M is the atomic weight, v_S is the sound velocity, and l is the MFP. $X = qc\hbar k_F/eH$, where q is the ultrasonic wave number, k_F is the Fermi radius, and e/c is the electronic charge. The quantity S_{11} was calculated on an electronic computer from the formula derived by Flax and Trivisonno.[3] This gives S_{11} in terms of power series in X and ql and is valid for all values of the arguments.

The curve-fitting procedure was similar to that used by Peverley.[4] For each set of experimental curves (attenuation versus field at several discrete temperatures) three parameters were used to obtain a fit to the data. The first, a scaling parameter for the abscissa, should give the numerical value of the quantity $qc\hbar k_F/e$. In choosing the second parameter, we elected to split off a factor ql from the first parenthesis in Eq. (1) and to define the scaling factor for the ordinate as that which would give the numerical value of $Nmv_F q/Mv_S$. The third parameter was then the quantity ql which determines the shape of the theoretical curve (attenuation versus field). Accordingly, in fitting our data we used the same scaling factors for each set of curves (which differed from each other only in the temperature) but allowed ql to vary from one curve to the next. In this way, we estimate that if the data were consistent with the CHH theory, we would have been able to assign values to the three parameters with uncertainties of not more than 1%. In fact, there were deviations from the CHH theory at high fields, and these introduced uncertainties into our values for $Nmv_F q/Mv_S$ and ql which varied from about 5% in the pure samples ($ql \sim 16$ at $T = 4.2$ °K) to about 20% in the less pure samples ($ql \sim 5$ at $T = 4.2$ °K).

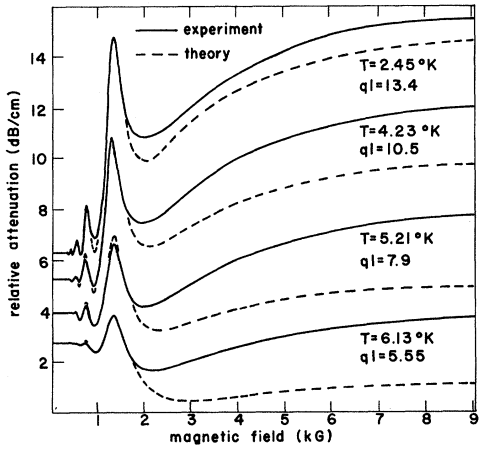

FIG. 1. Typical curves showing the comparison of data (———) with free-electron theory (- - - - - -) for various temperatures. The curves have been displaced on vertical scale for clarity.

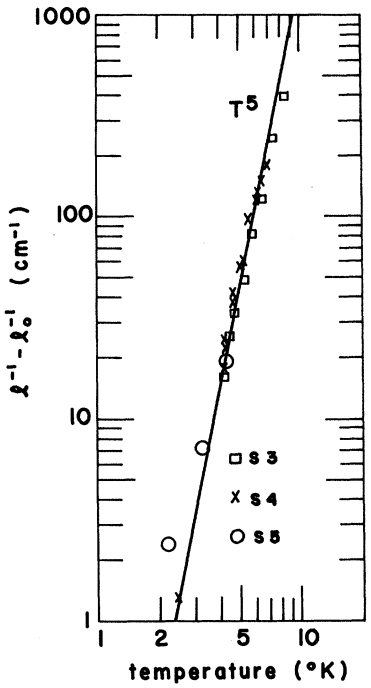

FIG. 2. Temperature dependence of the MFP as obtained parametrically from data on three samples. Comparison with a T^5 dependence is shown.

RESULTS

Temperature Dependence of MFP

Figure 1 shows typical fits of the theoretical curves to the data. The agreement is fairly good in the oscillatory portions of the curves where the magnetic field is small. From these fits a MFP was determined at each temperature. Fig. 2 is a plot of the temperature-dependent part of the MFP defined by $l^{-1} - l_0^{-1}$, where l_0 is the MFP extrapolated to $T = 0°$. The T^5 dependence of this quantity is in good agreement with the T^5 dependence of the ideal electrical resistivity predicted by the Bloch-Grüneisen law[5] and observed experimentally by MacDonald, White, and Woods[6] and by Natale and

Rudnick.[7] The absolute magnitudes also agree within experimental error (about 20% as determined from the scatter of points) under the assumption that the resistivity is related to the MFP by the expression

$$1/\rho = e^2 S l/12\pi^3\hbar\,, \qquad (2)$$

where S is the area of the Fermi surface.

Absolute Resistivities of Samples

These were determined at 4.2 °K by dividing the tabulated room-temperature value by the measured resistivity ratio. The resistivity was then converted into a MFP by means of Eq. (2). This MFP was invariably higher than that determined from the magnetoacoustic oscillations, by a factor which varied from 1.2 to 6 (for six samples) with an average of about 3. This result is consistent with an earlier observation by Peverley.[4] In making the resistivity ratio measurements (using the eddy current decay method of Bean, deBlois, and Nesbitt[8]) we verified that the strain introduced by the acoustic bond between sample and transducer did not affect the resistivity to any appreciable extent.

Scaling Factor for Ordinates

This quantity was converted into a numerical value for Nmv_F/M and compared with its theoretical value (under the assumption that $mv_F = \hbar k_F = 7.84 \times 10^{-20}$ gm cm/sec). The experimental value was smaller than the theoretical value, and the ratio (for five samples) varied from 0.58 to 0.96 with an average of 0.79. This would imply that the absolute attenuation is somewhat smaller than the theoretical attenuation, but we had no reliable method of measuring the absolute attenuation directly. Natale and Rudnick[7] claim to have made such absolute measurements in the low-frequency limit ($ql \ll 1$) and they found that the attenuation was about 1.6 times its theoretical value. In our experiments the ql ranged from 2 to 16.

Scaling Factor for Abscissa

This factor is determined essentially by fitting the periodicity of the magnetoacoustic oscillations and yields a value for the Fermi momentum. The values obtained were consistent with other measured values for this quantity, i.e., very close to the free-electron value, but do not represent an improvement in accuracy.

Attenuation at High Magnetic Fields

Invariably, it was found that the measured attenuation at high fields (i.e., beyond the oscillation region) was higher than the CHH prediction using the same parameters which optimized the low-field fit. This phenomenon was reported earlier by Peverley[4] and represents a qualitative deviation from the CHH theory. Since the latter (in our case) requires that the condition $\vec{q} \perp \vec{H}$ be satisfied, we verified that failure to satisfy this condition results in a deviation of the opposite sign to that actually observed. In considering possible causes for this effect we conclude that the relaxation-time assumption was the weakest link in the CHH argument; accordingly, we fitted our data by postulating an effective relaxation time and MFP which is defined so as to give agreement with the CHH theory. This relaxation time was higher at high magnetic fields than at low fields, the ratio varied from 1.1 to 1.25 with an average value of 1.2.

DISCUSSION

We first list the scattering processes which are likely to be significant in the temperature range of our experiments.

Normal Phonon Scattering (N processes)

The Bloch-Grüneisen law[5] which correctly predicts the T^5 dependence of the ideal resistivity at low temperatures is based on small-angle scattering from thermal phonons (the scattering angle is assumed to vary from zero up to T/Θ_D, where Θ_D is the Debye temperature, 90 °K for potassium).

Umklapp Scattering (U processes)

In the terminology of Young[9] there are 12 "hot spots" where the Fermi surface is closest to the zone boundaries and where the probability of Umklapp scattering is greatest. Hasegawa[10] has calculated the ideal resistivity of potassium on the basis of a realistic phonon spectrum and finds that the contribution from U processes exceeds that due to N processes at all temperatures above about 3 °K. Below this temperature, the U process contribution drops sharply. U processes are equivalent to a scattering angle in the neighborhood of 180°. Perhaps surprisingly, his theory predicts a temperature dependence of the ideal resistivity which is also close to T^5 in the range 3–8 °K and agrees well with the resistivity data of Natale and Rudnick.[7]

Impurity Scattering

Chambers[11] makes it clear that the relaxation-time assumption is equivalent to the assumption that the final state of ar electron after a collision process is independent of the initial state (in contrast to the situation for U processes or small-angle processes) and that this may be a fairly good description for large-angle scattering such as that due to impurities.

Small-Angle Scattering due to Dislocations, etc.

This type of scattering would be similar to that

caused by N processes but would persist down to absolute zero.

It should be remembered that, in considering the effectiveness of a given scattering process in returning the disturbed distribution of electrons to its equilibrium value, the form of the disturbed distribution function must also be taken into account. This quantity varies from one transport problem to the next. For example, in the case of d.c. electrical conduction, the form of the distribution function can be derived by assuming that the Fermi surface is displaced uniformly in the direction of the electric field.[12] For a spherical Fermi surface the disturbed distribution function (defined as the excess carrier density in phase space resulting from applied fields) varies as $\cos\theta$, where θ is the angle between a given point on the Fermi surface and the electric field direction. In the present context, it is of interest to consider the form of the disturbed distribution in the acoustic attenuation problem. For zero magnetic field, and in the limit $ql \gg 1$, this has been shown[13] to have a sharp peak in the equatorial belt of the (spherical) Fermi surface in the plane normal to $\vec{q}$. The electrons in that region remain in planes of constant acoustic phase and hence pick up more energy from the associated electric fields than less favorably situated electrons. If the peak in the distribution function is narrow, it would appear that a small-angle scattering process would be just as effective as a large-angle process in returning the distribution to equilibrium. On the other hand, when $ql \ll 1$, or alternatively, at high magnetic fields, all the electrons remain in regions of constant acoustic phase in the time interval between collisions, regardless of their position on the Fermi surface (perhaps not surprisingly, the acoustic attenuation in both these cases varies as q^2l, whereas it varies as q in the case $H = 0$, $ql \gg 1$). Although the form of the disturbed distribution function is not readily accessible from published theories of the magnetoacoustic effect, it is clear that there is no pronounced peaking effect when $ql \ll 1$ or at high magnetic fields and hence that in both cases, small-angle scattering processes will be less effective than large-angle processes (as in d.c. electrical conduction).

Thus, we appear to have a qualitative explanation for the apparent magnetic field dependence of the MFP, which is now interpreted in terms of the decreasing effectiveness of small-angle scattering processes as the magnetic field is increased.

If this interpretation is correct, further conclusions can be drawn. In our samples, the purity was such that scattering from thermal phonons and static defects were equal at about 5 °K. Hence, by varying the temperature, we could allow either category to become predominant. Since the high-field anomaly persisted over the whole temperature range (1.2–8 °K), it must be assumed that small-angle scattering from both N processes and from dislocations (and related defects) is important. However, we can dismiss the possibility that U processes are in some way wholly responsible for our observations because at the lowest temperature (~ 1.2 °K) they are almost certainly absent, according to Hasegawa's calculation.[10]

The magnetic field dependence of the effective MFP would suggest that small-angle scattering results in, at most, a 25% change in the effective MFP. This is consistent with the good agreement between our temperature-dependent MFP and the ideal resistivity data of Natale and Rudnick,[7] but can hardly account for the poor agreement between our acoustic MFP and our own resistivity data. This suggests that a much larger and more variable influence than small-angle scattering is present. One cannot exclude the possibility of systematic error in the resistivity measurements (which are notoriously difficult to avoid).

One ought to be able to predict, on the basis of our model, that the high-field discrepancy should vanish in the extreme limit $ql \gg 1$, when the physics of the problem becomes independent of the details of the scattering mechanism. Unfortunately, we were not able to obtain ql values higher than about 16, but even in this case the high-field fit was noticeably better. This may or may not be statistically significant.

CONCLUSION

The questions raised in this paper would probably be best resolved by further experiments on samples of higher purity and also by theoretical studies of the effects of small-angle scattering on the magnetoacoustic effect. At present, very few detailed treatments have been given of any transport problem other than with the conventional relaxation-time assumption. One such calculation was Pippard's[14] calculation of the conductivity of a free-electron gas for a transverse electric field of wave vector $\vec{q}$, using a simple diffusion model for small-angle scattering. The ql dependence of the conductivity differed at most by about 20% from the corresponding relaxation calculation. A numerical calculation along the lines of the CHH treatment but including a diffusion term of allow for small-angle scattering might well throw some light on the preceding discussion.

*Research supported by the U. S. Office of Naval Research, Contract Nos. 2249(03) and N00014-67A-0377-0007.

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PHYSICAL REVIEW B VOLUME 2, NUMBER 8 15 OCTOBER 1970

Modulated Piezoreflectance in Bismuth†

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Piezoreflectance has been measured in single crystals of bismuth with light polarized in the basal plane of the crystal for photon energies in the range 0.6 to 4.5 eV. The stress modulation of the optical reflectance was achieved by cementing a nearly circular disk of bismuth onto a lead-zirconate-titanate disk set into radial oscillations at 22 kcps. Using the Kramers-Kronig analysis, the induced changes in the imaginary part of the basal component of the dielectric tensor ϵ_{ij} have been computed from the piezoreflectance data. The energy dependence of the piezoreflectance $(\Delta R/R)_{11}$, as well as the computed changes in the dielectric constant $\Delta\epsilon_{11}'$, show an enhancement of the structure observed earlier by Cardona and Greenaway in the reflectivity spectrum. An assignment of the optical structure to direct interband transitions has been possible with the help of a comparison of the data with Golin's band-structure calculations.

INTRODUCTION

The electronic energy band structure of bismuth has been extensively investigated using the well-known experimental methods[1] of the de Haas–van Alphen effect, cyclotron resonance, magnetoacoustic attenuation, far-infrared absorption, magnetoreflection, and the various galvanomagnetic effects The results of all these experiments are in agreement with the two-overlapping-band model, [2–5] in which the Fermi surface for electrons in bismuth consists of three equivalent approximate ellipsoids in momentum space located at the symmetry points L in the Brillouin zone, and that for holes is a single ellipsoid of revolution located at the point T (Fig. 1). Using the two-band model, the experimental values of the direct energy gap at the point L, and the energy overlap between the conduction band and the valence band have been determined[4,6] to be $E_g = 15.3$ meV and $E_0 = 38.5$ meV, respectively. Starting with the Lin-Kleinman pseudopotential, [7] Golin[8] has determined its parameters for bismuth by adjusting to these experimentally determined values of E_g and E_0, and has then calculated the electronic energies of the various bands along several symmetry lines and planes. Although these calculations estimate the effective masses of electrons and holes to be approximately three times larger than the experimental values, the calculated direct interband energies are in agreement with the photon energies which give rise to peaks in the orbital reflectivity spectrum of bismuth.[9] Mase[10] and Ferreira[11] have also calculated the band structure of bismuth using the tight-binding and the augmented-plane-wave (APW) methods, respectively.

Recently interband transitions in solids have been determined by means of electric field, [12–14] temperature, [15,16] and stress[17–19] modulation of the optical reflectivity and absorption. It is well known now that the structure in the photon energy dependence of the imaginary part of the dielectric constant is considerably enhanced by such modulation