Magnetoacoustic Study of the Mean Free Path of Electrons in Cadmium and Zinc

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(Received 26 July 1965)

Using a free-electron model, the temperature and angular variations of the electron mean free path in cadmium and zinc have been determined from magnetoacoustic experiments. The mean-free-path dependence of the high-field saturation attenuation and the ultrasonic open-orbit resonance height were used for investigations in cadmium from 1 to 9°K and in zinc from 1 to 4.2°K. When the data were corrected for the temperature variation of the Debye temperature, it was found that the mean free path varies approximately as T^{-5} for electrons in closed orbits and as T^{-4} for electrons in open orbits. Anomalous behavior in the temperature variation is observed below 3°K and seems to be related to the resistance-minimum effect. The interpretation of the angular variation of the mean free path is complicated because cadmium and zinc have Fermi surfaces of more than one sheet, but the data indicate approximately an order-of-magnitude variation over the Fermi surface in both cadmium and zinc.

I. INTRODUCTION

PREVIOUS investigations have shown that various magnetoacoustic techniques can provide information about the mean free path l of electrons in pure, single-crystal materials.[1,2] The present study is an analysis of the variation of the mean free path of electrons in cadmium and zinc, using two methods: (1) the high-magnetic-field attenuation of compressional ultrasonic waves for different crystal directions and temperatures; and (2) the temperature dependence of the ultrasonic open-orbit resonance phenomena.[3] The cadmium and zinc single crystals were of such purity that the electron scattering was predominantly by thermal phonons above about 3°K. Measurements of the high field attenuation and open-orbit resonance in cadmium were made over the temperature range from 1.0 to 9.0°K, while the data on zinc were confined to temperatures from 1.0 to 4.2°K.

The relationship between the high-field-attenuation saturation value and the mean free path, using the free-electron model, is evident from the expression derived by Pippard[4] for the compressional attenuation α_L in the limit as the magnetic field $H \to \infty$,

$$\lim_{H\to\infty} \alpha_L(H) = N m_0 v q^2 l / 15 \rho v_s. \quad (1)$$

Here N is the number of free electrons per unit volume, m_0 the electron mass, v the Fermi velocity, q the magnitude of the sound wave vector ($q=2\pi\nu/v_s$), ν the sound frequency, v_s the sound velocity, and ρ the metal density. It is seen that the limit of the attenuation in high magnetic fields is proportional to q^2l, and it has been shown[2] that this limit is approached as H^{-2}.

Experimentally, the difference between $\alpha_L(H)$ and the zero-field attenuation α_0 is usually determined. A numerical calculation[2] shows that the high-field saturation attenuation is equal to the attenuation in zero field for $ql=6.8$, thus allowing a rather direct experimental determination of l using the free-electron model. The mean free path determined involves an average over that part of the Fermi surface being traversed. Different orbits can be selected by changing the configuration of sound and field directions, and it should therefore be possible to establish the anisotropy of the mean free path over the Fermi surface. The results of such a procedure are much more difficult to interpret for metals with Fermi surfaces of several sheets and can be complicated by real metal effects and the presence of the Fermi velocity v in the expression for the high-field attenuation. These difficulties of interpretation are not as severe in measurements involving only the temperature variation of $\alpha_L(H\to\infty)$ for a particular sound and field configuration.

That information concerning the mean free path of open-orbit electrons can be obtained by ultrasonic studies has been discussed previously.[3] The ultrasonic open-orbit resonance has a Lorentz shape and at the resonance maximum the attenuation is found to be[5]

$$\alpha_m = \alpha_0 q l / \pi m, \quad (2)$$

where m is the resonance order and α_0 the attenuation in zero magnetic field. For the case $ql \gg 1$, the zero-field attenuation[4] α_0 is proportional to q and independent of l, so that α_m is proportional to q^2l. In the limit of $ql \ll 1$, α_0 varies as q^2l so that α_m is proportional to q^3l^2. The resonance width is found to be inversely proportional to l, but determination of the variation of l from the width is not nearly as straightforward as is the measurement of α_m. The ultrasonic resonance effect in cadmium and zinc is caused by electrons moving essentially parallel to [0001] (in momentum space) on the arms of the second-band hole surface.[6,7] The resonance is seen for sound propagation **q** along either [10$\bar{1}$0] or [$\bar{1}$2$\bar{1}$0]

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with magnetic field **H** mutually perpendicular to [0001] and **q**. Measurement of the temperature variation of α_m at a fixed frequency thus allows a direct determination of the temperature dependence of l for this select group of electrons.

II. EXPERIMENTAL

The pulsed ultrasonic experiments were performed in the customary manner, using frequencies from 10 to 70 Mc/sec. Magnetic fields up to 19 000 G were utilized, the field values being obtained from a rotating-coil gaussmeter which was calibrated by nuclear-magnetic-resonance equipment. The cadmium and zinc single crystals used in the measurements were cut so that sound could be propagated between a pair of [$\bar{1}2\bar{1}0$] or [$10\bar{1}0$] faces.

Temperatures below 4.2°K were obtained by pumping on the liquid-helium bath, the temperature being determined by measuring the vapor pressure of the bath. For temperatures between 4.2 and 9.0°K, the sample chamber was evacuated at various pumping rates with sufficient heat being added to warm the sample to the desired temperature. Above 4.2°K the temperatures were measured with a nominal 1000-ohm carbon-resistance thermometer which was calibrated below 4.2°K at several liquid-helium points and ultrasonically above 4.2°K at the superconducting transition points of high-purity lead and niobium single crystals. A superconducting wire calibration element was also positioned in good thermal contact with the sample platform and was used as an internal check on the carbon resistor. This element consisted of lengths of Ta, Pb, and NbZr wires connected in series, the resistance of the element being monitored during each temperature run. The superconducting-to-normal resistive transitions served as excellent temperature indicators. Temperatures are believed accurate to ±0.01°K below 4.2°K and ±0.05°K above 4.2°K. It should be mentioned that the ultrasonic seals between the sample and transducer were made with Dow Corning High Vacuum Grease.

III. RESULTS

A. Temperature Variation of l

The first step in the analysis of the high-field-attenuation data is to determine the saturation attenuation $\alpha=\alpha(H\rightarrow\infty)$-$\alpha_0(H=0)$. This was done by plotting the relative attenuation values at high fields against H^{-2} and extrapolating to infinite field. Data at a number of temperatures were taken for sound propagation along [$10\bar{1}0$] and [$\bar{1}2\bar{1}0$] in transverse magnetic fields.

In Fig. 1 is shown the temperature variation of α for cadmium at 72 Mc/sec for **q**∥[$10\bar{1}0$] and **H** at various angles relative to [$\bar{1}2\bar{1}0$]. A similar plot for q∥[$\bar{1}2\bar{1}0$] at 31 Mc/sec is shown in Fig. 2. The dashed lines are for illustrative purposes and do not represent the temperature fit described below. It should be pointed out that some uncertainty exists in using the present analysis for the open-orbit configurations, since the high-field saturation behavior of open orbits is different from that of closed orbits.[8] Since there is no way at present of determining how much of the high-field attenuation is caused by the open orbits, the data were analyzed in the same manner as for closed orbits. The data points shown in Figs. 1 and 2 were analytically fit to curves of the form

$$\alpha(l)=A+BT^{-n}, \tag{3}$$

where n was determined from logarithmic plots of $\alpha(l)$ as a function of T. It was possible to fit the data suitably to equations of this type at temperatures above the anomalous behavior (to be discussed later) up to the temperature where $ql\sim8$, i.e., $l\sim1.3\lambda$, where λ is the ultrasonic wavelength. Values of A, B, and n obtained in this manner are given in Table I. It is seen that the attenuation, and therefore the mean free path, shows an approximate T^{-4} dependence in the temperature range studied. The temperature dependence varies

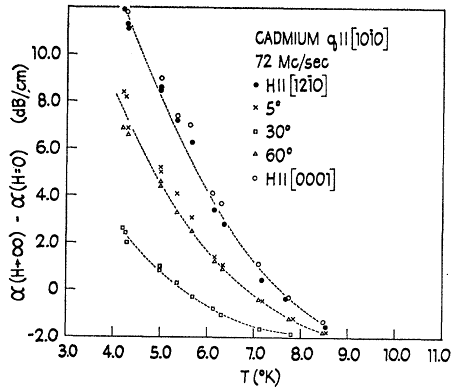

FIG. 1. Temperature variation of $\alpha(H\rightarrow\infty)$-$\alpha_0(H=0)$ in cadmium for 72 Mc/sec sound propagated along [$10\bar{1}0$] with magnetic field at various angles relative to **H**∥[$\bar{1}2\bar{1}0$].

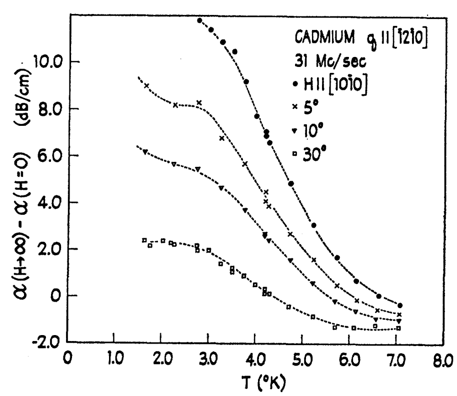

FIG. 2. Temperature variation of $\alpha(H\rightarrow\infty)$-$\alpha_0(H=0)$ in cadmium for 31 Mc/sec sound propagated along [$\bar{1}2\bar{1}0$] with magnetic field at various angles relative to **H**∥[$10\bar{1}0$].

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TABLE I. Constants determined by fitting the temperature variation of the attenuation to curves of the form $\alpha = A + BT^{-n}$ for cadmium. The error limits on A and B determined from the fitting procedure are $\pm 5\%$.

Configuration		Frequency Mc/sec	A (dB/cm)	B	n (± 0.2)
	$\mathbf{H}\|[\bar{1}2\bar{1}0]$	72	1	2100	3.6
$\mathbf{q}\|[10\bar{1}0]$	5°	72	0	1400	3.6
$\alpha_L(H\rightarrow\infty)$	30°	72	0	340	3.6
	60°	72	0	1400	3.6
	$\mathbf{H}\|[0001]$	72	1	2100	3.6
	$\mathbf{H}\|[10\bar{1}0]$	31	0.6	2000	4.0
	5°	31	0	1300	4.0
$\mathbf{q}\|[\bar{1}2\bar{1}0]$	10°	31	−0.6	970	4.0
$\alpha_L(H\rightarrow\infty)$	30°	51	0.5	890	4.0
	60°	51	−0.3	340	4.0
	$\mathbf{H}\|[0001]$	51	−0.3	340	4.0
Open-orbit resonance height	$\mathbf{q}\|[10\bar{1}0]$ $\mathbf{H}\|[\bar{1}2\bar{1}0]$	72	1	540	3.0
	$\mathbf{q}\|[\bar{1}2\bar{1}0]$ $\mathbf{H}\|[10\bar{1}0]$	31	−1	310	3.0

somewhat for different configurations or sets of orbits on the Fermi surface.

The temperature variation of the open-orbit resonance was deduced from x-y recordings of attenuation as a function of magnetic field. Typical recorder tracings are shown in Fig. 3 for cadmium with $\mathbf{q}\|[10\bar{1}0]$ and $\mathbf{H}\|[\bar{1}2\bar{1}0]$. The normal magnetoacoustic oscillations occur up to about 500 G and the open-orbit resonance occurs at 1350 G at 72 Mc/sec. The quantity α_m is the height of the resonance maximum relative to $\alpha_0(H=0)$ and was determined by a precise voltage measurement. The α_m data are shown in Fig. 4 for cadmium for $\mathbf{q}\|[10\bar{1}0]$ at 72 Mc/sec and $\mathbf{q}\|[\bar{1}2\bar{1}0]$ at 31 Mc/sec. The experimental points were fit to Eq. (3) as before, and the constants are given in Table I. The temperature variation of the mean free path of the electrons moving in open trajectories is seen to exhibit approximate T^{-3} behavior.

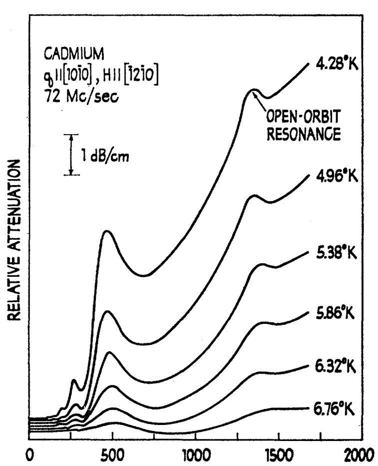

FIG. 3. Recorder tracings of the temperature variation of the relative attenuation of 72 Mc/sec sound in cadmium for sound along $[10\bar{1}0]$ and field along $[\bar{1}2\bar{1}0]$.

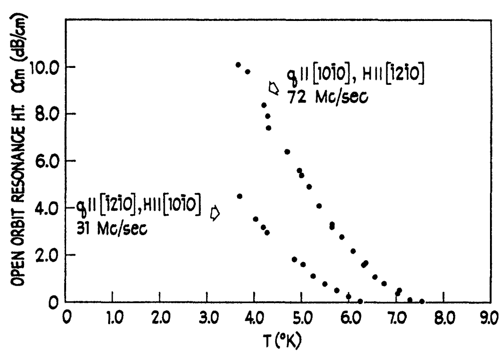

FIG. 4. Height of the open-orbit resonance relative to $H=0$ for the two open-orbit configurations studied.

B. Anomalous Behavior

In a previous publication, anomalous behavior of the temperature variation of the electron mean free path in cadmium was reported.[9] It was found that the l-versus-T curves exhibited maxima and minima around 3°K. This phenomenon has been observed in three crystals of cadmium and one crystal of zinc, and is reflected in the temperature variation of the high-field attenuation as well as the height of the open-orbit resonance.

In Fig. 5 are shown some data taken below 4.2°K for both cadmium and zinc, showing the temperature dependence of l for various sound directions and field angles. These data were analyzed as described previously[2] and therefore yield directly the temperature dependence of the average mean free path for the particular sound and field configuration under consideration. It is seen from Fig. 5 that maxima and minima occur in both cadmium and zinc in the range from 2.5 to 3.5°K. The effect seems to be most pronounced for $\mathbf{q}$ along $[10\bar{1}0]$ and is stronger in cadmium than in zinc. This anomalous behavior is very similar to the resistance minimum effect reported widely over the

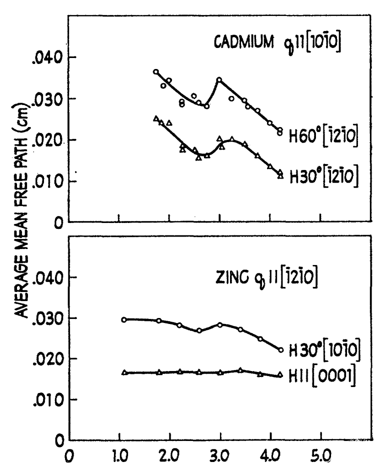

FIG. 5. Average electron mean free path determined from high-field acoustic-attenuation data as a function of temperature for cadmium and zinc showing anomalous behavior.

[9] B. C. Deaton, Phys. Letters **7**, 7 (1963).

past several years.[10–15] The resistance minimum is apparently caused by the presence of certain magnetic impurities which introduce a temperature-dependent term to the resistance.[16–18] This term involves spin-dependent scattering at the Fermi surface by the magnetic moment of the impurities. Spectrographic analysis of the cadmium sample used in the present measurements indicated major impurities of a few parts per million of iron and lead. The results of the spectrographic analysis, plus the close similarity of the present data to previous resistance-minima measurements, are quite indicative that the effect shown in Fig. 5 is just the behavior expected if the cadmium and zinc show a resistance minimum. From the data of Hedgecock and Muir[14] on zinc and Muir[15] on cadmium containing small amounts of manganese, it is found that the resistance minima should occur below about 5°K for purities equal to those of the present samples. It therefore appears that for extremely pure crystals, magnetic impurities in concentrations of a few parts per million can have definite effects on the transport properties.

C. Angular Anisotropy

The angular anisotropy of the high-field-limiting attenuation has been used to provide information concerning the anisotropy of the relaxation time τ ($v\tau=l$) in copper.[2] In the cases of cadmium and zinc, however, an analysis of this kind is more complicated for several reasons. In the first place, cadmium and zinc have Fermi surfaces consisting of more than one sheet and therefore for most sound and field configurations several quite different pieces of the surface may contribute to the attenuation. Secondly, as mentioned earlier, the high-field attenuation could reflect changes in the Fermi velocity (i.e., a property of the Fermi surface) as well as changes in mean free path. In addition, the presence of open orbits for certain directions further complicates the investigation. Disregarding these difficulties for the time being, the high-field data can be reduced as described earlier, an average Fermi velocity assumed, and the angular variation of an average mean free path found. Data on cadmium and zinc are shown in Fig. 6. These data are essentially equivalent to ordinary high-field polar plots except for the present extrapolation to infinite field. As is seen in Fig. 6, curves at 4.2°K and curves at lower temperatures, which represent entirely different regions of scattering, have essentially the same shape. It seems

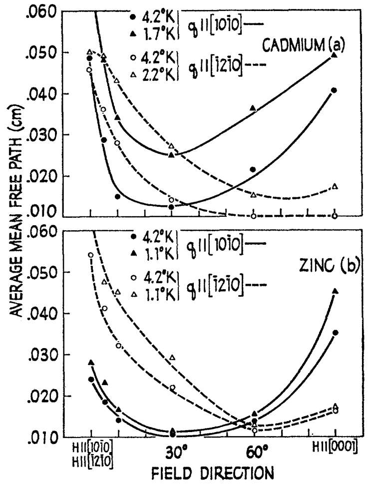

FIG. 6. Angular anisotropy of the mean free path determined from high-field acoustic-attenuation data for cadmium and zinc. All data were taken in transverse magnetic fields.

probable that scattering by impurities would show a different anisotropy from thermal scattering, so that quite differently shaped curves might be expected for the two regions. The fact that there is very little difference in the scattering anisotropy indicates that the high-field attenuation anisotropy in the present cases may be caused in part by the Fermi velocity. Without further theoretical investigations of which parts of the Fermi surface contribute to the high-field limiting attenuation, there seems little hope of a complete solution for Fermi surfaces as complicated as those of cadmium and zinc.

IV. CONCLUSIONS

The electrical conductivity of a metal is found to be[19]

$$\sigma=N_{\rm eff}e^2l/m_0v\,, \tag{4}$$

where $N_{\rm eff}$ is the effective number of free electrons. At temperatures below the Debye temperature Θ, taking into account the density of phonons available for scattering of electrons and the fact that small angle scattering is required by conservation of momentum, it is found that σ increases[19] as $(\Theta/T)^5$. This T^{-5} behavior is indeed found experimentally in high-purity cadmium and zinc single-crystal wires.[20] Since l is related directly to the electrical conductivity, an ultrasonic determination of the temperature dependence of l would also be expected to exhibit T^{-5} behavior. Several factors might be expected to cause the present raw data to disagree somewhat with a normal T^{-5} dependence: (1) Any temperature dependence of the Debye temperature in this range would be reflected directly in the present data, since l should vary as $(\Theta/T)^5$; (2) the possibility that the mean free path governing ultrasonic processes

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is different from the resistive mean free path as suggested by Steinberg,[21] Filson,[22] and Lax[23]; and (3) the fact that the present studies measure l in a magnetic field, whereas the T^{-5} dependence is for a field-free case.

Recent low-temperature measurements of the heat capacity of cadmium by Phillips[24] provide a determination of the temperature variation of Θ. The present results can therefore be corrected for the variation of Θ in the temperature range studied. The data of Phillips indicate that the Debye temperature Θ varies approximately as $T^{-0.26}$ so that the temperature variations given in Table I should be multiplied by a factor $(T^{-0.26})^5=T^{-1.3}$. It is seen, therefore, that this correction brings the present results on the temperature variation of l into very close agreement with a T^{-5} law. The results of this investigation therefore suggest that there may not be such a great difference between acoustically and resistively measured mean free paths. This same conclusion was reached in a study of the absolute electronic attenuation of compressional waves in copper by Kolouch and McCarthy.[25]

It is of interest to note that the temperature dependence of l of the open-orbit electrons is somewhat different from those in closed orbits. When the data of Table I are corrected for the Debye temperature variation, it is seen that l varies roughly as T^{-4} for the open-orbit electrons. The reason for the observed difference is not known, but it is probably a real effect related to the character of this select group of electrons.

The anomalous behavior of l observed in both cadmium and zinc indicates that transport properties in high-purity materials can definitely be influenced by small amounts of magnetic impurities. The temperature of the mean-free-path maximum in our data seems to decrease with purity (i.e., value of l for the particular orbit or sample involved) as is expected.[14]

Determination of the anisotropy of the mean free path (or relaxation time) on the Fermi surfaces of cadmium and zinc by using the present technique is complicated by the previously mentioned factors. If our data are analyzed in the usual manner, it is found that the mean free path varies by as much as approximately a factor of six over the Fermi surfaces of cadmium and zinc. This is in good agreement with the estimation given by Gibbons and Falicov.[26] The angular variation of l is found not to differ greatly for different regions of scattering, i.e., the region where electron scattering is predominantly by impurities at about 1 °K, and the region where thermal phonon scattering is dominant near 4 °K. This fact suggests that the angular dependence might reflect Fermi-surface properties instead of mean-free-path anisotropy, at least in the present cases.

In conclusion it should be noted that the high-field saturation technique based on a free-electron model appears to afford an excellent method of investigation of electronic free paths in pure bulk materials. It proves to be quite easy to find the frequency at which the high-field attenuation is equal to that in zero field, and it is felt that this technique is considerably more accurate than estimating ql from the number of magnetoacoustic oscillations obtained.[2] Recent investigations[27] in Pb show that the high-field technique is useful also in determining ql at various frequencies and temperatures for a superconductor, assuming the attenuation at the critical field to be the comparison base.

ACKNOWLEDGMENTS

The author gratefully acknowledges helpful discussions with J. D. Gavenda, J. W. Hagemeyer, F. A. Blum, Jr., and K. B. Ward, Jr.

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