Size effects in the magnetoresistance of rolled potassium films

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The magnetomorphic effects of rolled potassium films are reported for a sample temperature of 4.2 K with the magnetic field parallel to the sample surface. The film thickness is between 1.8 and 108 μm. With the magnetic field perpendicular to the current, there is a resistance maximum. The product, $\omega\tau$, of the cyclotron frequency and the scattering relaxation time at the maximum for different film thicknesses does not agree with predictions of free-electron theory and is presented as an empirical relation. The transverse magnetoresistance was studied with fields up to $\omega\tau\sim 32$. We found that the development of a size-effect saturation pattern is a function of the ratio of the film thickness to the mean free path. The magnetoresistance in the longitudinal geometry has a broad maximum and tends towards saturation at high fields up to $\omega\tau=140$. For both the transverse and longitudinal geometries there is only a qualitative agreement between our observations and available theories of size effects for a free-electron metal.

I. INTRODUCTION

The observation of size effects in the resistivity of thin metallic films can be traced back to the 19th century.[1] The first major theoretical impact was provided by Fuchs[2] who developed an exact solution of the influence of size effects upon the electrical conductivity in a free-electron metal. The effect of the application of a magnetic field to a metallic specimen with a characteristic dimension similar to the electronic mean free path was studied by Sondheimer,[3] MacDonald,[4] Dingle,[5] Chambers,[6] Olsen,[7] Azbel',[8] Gurevich,[9] and many other scientists[10] who predicted theoretically and established experimentally a great variety of magnetomorphic phenomena. Although a great deal of study has been done on different metals which are far from being "free-electron" ones, it happened, somehow, that the alkali metals, the best candidates for checking the size-effect models based essentially on the free-electron approximation, are investigated the least.[11] In our literature search we found that the predictions of the magnetoresistance behavior in the MacDonald[4] geometry, were checked in early work with sodium by MacDonald and Sarginson,[12] Chambers,[6] and White and Woods.[13] Taub and coworkers[14] studied the transverse magnetoresistance in relatively thick potassium wires ($\sim$2 mm) and found some weak evidence of size effects, which they were unable to analyze completely. The oscillatory effects from a magnetomorphic origin predicted by Sondheimer[3] for thin metallic films have been reported for sodium wires by Babiskin and Siebenmann.[15] A search for size effects in the Hall voltage was done at 90 K by Cirkler[16] who studied evaporated potassium films of very small thickness. There are no reports on the observation of these phenomena in potassium films with an intermediate thickness (tens of μm). This issue is the main concern of the present paper. It should be noted that investigation of size effects in potassium is also of fundamental importance for checking the conceptual grounds underlying the more complicated modern theories of condensed matter.

In the MacDonald geometry, the magnetic field is perpendicular to the current direction and in the plane of the film. The predicted magnetic-field dependence of the resistivity in this geometry determined from free-electron theory[12,17] is illustrated in Fig. 1. The variable $\beta=d/r$, where d is the film thickness and r is the cyclotron radius, is proportional to the magnetic field B, since $r=mvc/eB$. There is an initial increase at low magnetic-field strength because some electrons suffer an initial decrease in their free path.[4] There is a maximum at a magnetic field corresponding to $\beta=0.55$. In larger magnetic fields, the resis-

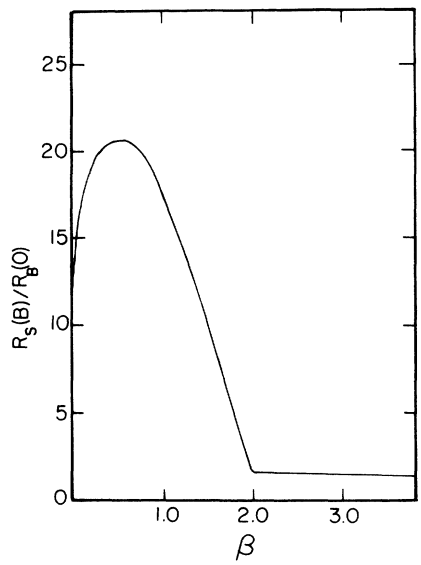

FIG. 1. Schematically drawn qualitative behavior of the ratio $R_S(B)/R_B(0)$, where $R_S(B)$ is a field-dependent resistivity of the sample, influenced by the presence of size effects, and $R_B(0)$ is the zero-field bulk resistivity of the sample material, both measured at $T=4.2$ K (based on Fig. 3 of Ref. 17). The variable $\beta=d/r$ (where d is the film thickness and r is the maximum electronic orbit radius) is linearly proportional to the field strength B.

tivity decreases with increasing field beyond the maximum because the effective electron free path of the spiral motion in the magnetic field is increased by reducing collisions with the surface. For $\beta > 2.0$ when the film thickness is greater than the diameter of the cyclotron orbit, the resistivity is expected from free-electron theory to be independent of magnetic field and to show saturation of size effects. However, measurements with sodium wires[4,13] show that the resistance slightly increases with field in this region. This increase has been attributed[4,13] to bulk magnetoresistance.

The method of preparing potassium films by a rolling technique is described in Sec. II. They were rolled reliably to any thickness in the μm range because of the very soft nature of potassium and were annealed at room temperature to reduce strain. All the samples provided reproducible results which are described in Sec. III. The resistivity data were taken with the magnetic field parallel to the surface of the film at a sample temperature of 4.2 K. In the MacDonald geometry with the magnetic field perpendicular to the current direction, the magnetic-field range was as high as 1.8 T. In the longitudinal geometry with the magnetic field parallel to the current direction, magnetic fields up to 7.8 T were used. Characteristic features of the magnetic-field dependence of the resistivity are analyzed in Sec. IV and predictions of the available theory developed for free electrons are tested carefully. It is concluded in Sec. V that this theory is not satisfactory for a quantitative explanation of the magnetic-field dependence of the resistivity of the films.

II. EXPERIMENTAL

We now give a brief description of the sample preparation procedure, which is quite important when one deals with potassium because of its high chemical activity and the frequently reported observation of a poor reproducibility of the experiments performed with this metal.[14] We found, however, that the study of size effects is reproducible, with the use of freshly cut, unoiled potassium for the film preparation. At 4.2 K, the electronic mean free path of pure bulk potassium calculated in a free-electron model is as large as ~ 80 μm. In order to prepare thin potassium samples with the thickness of tends of μm one can simply roll the films from a small piece of metal and allow them to thermally anneal before cooling them to 4.2 K. The films were prepared from potassium metal held in a glass ampoule and manufactured by the Callery Chemical Company.[18] The preparation was done in a dry box filled with dry nitrogen gas. A small amount of potassium was taken from the glass ampoule and placed on a machined, Kel-F substrate with the shape recommended by Hurd[19] for performing Hall-effect studies.[20] The in-plane dimensions of the Kel-F substrate, length (L) and width (W), were the same for all our samples with $L=3.84$ cm and $W=0.41$ cm. Using the fact that the fresh, unoiled potassium wets Kel-F, the small amount of metal was spread and rolled with a stainless-steel roller on the substrate with an average thickness down to 1.8 μm. The shape of the sample was cut to that of the substrate. The sample thickness was determined three hours after the film preparation from the room-temperature four-probe resistance using the bulk resistivity of pure potassium. The sample was put in a brass cell that was closed with an indium seal. From the cell there was an electrical feedthrough and a tube closed with a valve. The cell was refilled (after it was removed from the dry box) with pure He gas, transferred through a liquid-nitrogen cold trap.

Six electrical contacts to the sample were made with thin, silver-coated wires oriented perpendicular to the longest axis (L) and mechanically pressed towards the film with a Kel-F plate. These reliable wire contacts reduced possible point stresses and nonlaminarity of the current along the longest sample dimension.

The resistivity was measured by the four-probe method with a computer control. The direction of the current with a magnitude of 10 mA was switched at 5-sec intervals. The difference in the average potential for the current in the two directions for pairs of measurements provided the potential measurement with a sensitivity of better than 10 nV and eliminated thermal voltages. The potential was measured with a Keithley nanovoltmeter, Model 149. The direction of the magnetic field parallel to the film surface was reversed to eliminate the influence of the Hall field on the resistance measurements. The magnetic field was provided by an electromagnet with a maximum field of 1.8 T for measurements in the MacDonald geometry and by an 8.5-T superconducting solenoid for the longitudinal geometry with the magnetic field parallel to the current direction.

The orientation of the magnetic field B is defined with respect to the outward normal to the film's surface. The magnetic field was aligned parallel to the sample surface ($\theta=90°$) in the following way. First, the direction perpendicular to the surface was determined from the maximum Hall effect. This direction, we note, was found to give the maximum linear slope of $\Delta R(B)/R(0)$ versus B at 1.8 T. Measurements were then taken 90° from this direction to be in the MacDonald geometry with the magnetic field in the film plane and perpendicular to the current flow.

The residual-resistance ratio of the bulk material is

$$\mathcal{R}_B = \frac{R_B(295)}{R_B(4.2)} , \tag{1}$$

where $R_B(295)$ and $R_B(4.2)$ are the resistances at 295 and 4.2 K, respectively. The residual-resistance ratio of a sample that exhibits size effects is

$$\mathcal{R}_S = \frac{R_S(295)}{R_S(4.2)} . \tag{2}$$

Equations (1) and (2) are related by the approximate Nordheim form of the Matthiessen rule[6] for diffuse scattering by

$$\mathcal{R}_B = \mathcal{R}_S \left[1 + \frac{1}{\kappa} \right] , \tag{3}$$

where $\kappa = d/l$ for a bulk mean free path l with a film thickness d.

The product, $\omega\tau$, of the cyclotron frequency and the

scattering relaxation time is given from the free-electron expressions as

$$\omega\tau=\frac{eB\tau}{mc}=\frac{R_H}{\rho_B(4.2)}B \ , \tag{4}$$

where R_H is the Hall constant and ρ_B is the bulk resistivity. Then from Eq. (1)

$$\omega\tau=\frac{R_H}{\rho_B(295)}\mathcal{R}_B B \ . \tag{5}$$

In terms of the size-effect residual-resistivity ratio this becomes from Eq. (3)

$$\omega\tau=\frac{R_H}{\rho_B(295)}\mathcal{R}_S\left[1+\frac{1}{\kappa}\right]B \ . \tag{6}$$

With the known values of R_H and $\rho_B(295)$

$$\omega\tau=6.19\times10^{-3}\mathcal{R}_S\left[1+\frac{1}{\kappa}\right]B \ , \tag{7}$$

where the magnetic field is expressed in tesla. Equation (7) is used in this work for the magnetic field parameter to plot $\Delta R_S(B)/R_S(0)=[R_S(B)-R_S(0)]/R_S(0)$.

The bulk resistance $R_B(B)$ in a magnetic field B can be written in the Kohler form[21]

$$\frac{\Delta R_B(B)}{R_B(0)}=f(B/\rho_B) \ . \tag{8}$$

This becomes $f(\omega\tau)$ from Eq. (4). This is generalized by Olsen[7] to the case of magnetoresistance of samples with size effects as

$$\frac{\Delta R_S(B)}{R_S(0)}=f_S(\omega\tau) \ , \tag{9}$$

with the subscript S on the right-hand side indicating that the $\omega\tau$ is calculated from Eq. (7).

The values of $\omega\tau$ calculated from Eq. (7) should be similar for different samples prepared from the same material. This is valid for our samples as shown in Table I which gives $\omega\tau$ at 1.8 T for the different samples. The value for sample 1, which was prepared from material that had been immersed in oil, is a little lower. All the other samples were prepared without oil. Table I also gives the room-temperature resistance and the residual-resistance ratio of our samples. The average thickness, determined from the room-temperature resistance, was between 1.8 and 108 μm. The values of κ were determined from Eq. (3) and correspond to a range of l between ~30 and ~80 μm.

III. RESULTS

The magnetoresistance in the MacDonald geometry of sample 4 with a thickness of 1.8 μm (Fig. 2) has a maximum at $\omega\tau=5$ and then decreases with increasing field up to the maximum $\omega\tau$ of ~32. We stress that the results for samples 2 and 4 with similar κ are essentially alike and indicate excellent reproducibility of the results from the same method of sample preparation.

The magnetoresistance for a sample thickness of 20.9 μm (sample 3) shown in Fig. 3 has a maximum at $\omega\tau\sim2$ and essentially saturates above $\omega\tau=20$. Samples 1 and 3 have the same thickness. However, sample 1 was prepared from potassium metal stored in organic oil which was cleaned off before the sample preparation. There was still a very thin oil layer on top of the sample after it was rolled to the desired thickness. This significantly affected the size-effect data and drastically influenced the reproducibility of the results. The maximum in $\Delta R_S(B)/R_S(0)$, which is clearly seen for sample 3 in Fig. 3, is absent for sample 1. Also, sample 1 did not stand up to thermal cycling and four of the six electrical contacts to the sample were lost after two warming-cooling cycles.

Results for the thickest sample ($d=108$ μm) are shown in Fig. 4. There is a maximum at low fields and a minimum at $\omega\tau=8$. The magnetoresistance increases with magnetic field above $\omega\tau=10$ with a Kohler slope of 6.3×10^{-3}.

While there is no minimum in the magnetoresistance in the strict MacDonald geometry in magnetic fields up to $\omega\tau=32$ for $d\leq21$ μm it does appear for all sample thicknesses when the magnetic field is inclined to the sample surface. This is shown for a direction 15° from the surface of sample 3 in Fig. 5. There is a minimum at $\omega\tau=14-15$ as well as a maximum at $\omega\tau=2.3$. The Kohler slope at high fields is 1.2×10^{-2}.

The magnetoresistance with the magnetic field parallel to the sample surface for sample 2 and current direction (the longitudinal geometry) is shown in Fig. 6 for magnetic fields up to 7.8 T corresponding to an $\omega\tau$ of 140. Here there is a maximum at $\omega\tau=12$ and there appears to be saturation at high magnetic fields.

TABLE I. Various parameters of the film samples studied in this work. Here κ is a ratio at film thickness to mean free path; $\mathcal{R}_S$ is a residual-resistance ratio for film samples; $\omega\tau$ is evaluated from Eq. (7) at $B=1.8$ T.

Sample	R_S(295 K) ($\mu\Omega$)	$\mathcal{R}_S$	d (μm)	κ	$\omega\tau$
1	31.7	537	21.0	0.259	29.1
2	347.0	96	1.9	0.034	32.6
3	32.0	601	20.9	0.258	32.7
4	363.7	150	1.8	0.055	32.1
5	6.2	1562	108.0	1.333	30.5

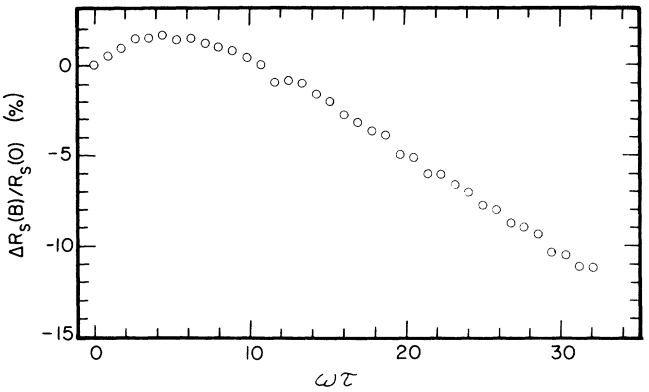

FIG. 2. The transverse magnetoresistance of sample 4 in the MacDonald geometry.

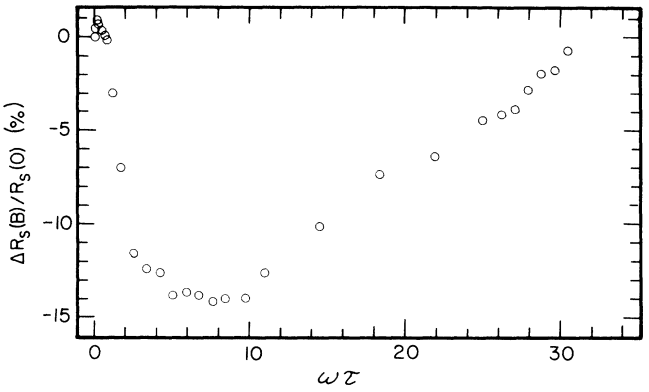

FIG. 4. The transverse magnetoresistance of sample 5 in the MacDonald geometry.

IV. DISCUSSION

Figures 2–4 show the development of the MacDonald size effect as a function of κ, the ratio of the film thickness to the mean free path. As κ increases from 0.055 to 1.33 in Figs. 2–4, the MacDonald maximum appears at lower values of $\omega\tau$. At high fields, increasing κ results in the development from a linear decrease of $\Delta R_S(B)/R_S(0)$ in Fig. 2, through saturation in Fig. 3 to a well-resolved minimum in Fig. 4. According to the theoretical predictions the onset of the saturation in size effects should occur at $\beta\sim 2$ (or $\omega\tau\sim 2/\kappa$) which gives for samples 2, 4, 3, and 5 the $\omega\tau=59$, 36, 8, and 1.5, respectively. One sees that with the maximum available $\omega\tau\sim 32$ samples 2 and 4 do not show any saturation. However, for the thicker samples 3 and 5 the onset of saturation occurs at much higher $\omega\tau$ than provided by the theory.

The initial maximum should be independent of κ and is expected to appear at $\beta=d/r=0.55$ according to Ditlefsen and Lothe.[17] This condition requires that the $\omega\tau$ at the maximum is

$$\omega\tau(\max)=\frac{0.55}{\kappa} . \tag{10}$$

This relation is tested in Fig. 7(a) where $\omega\tau$(max) is plotted as a function of $1/\kappa$. There is a good agreement between the prediction of Eq. (10) and the results for the two thickest samples but not for the thinner samples. We found, however, that the observed $\omega\tau$(max) can be fit for all samples by the empirical relation

$$\omega\tau(\max)=\exp(-3.4\sqrt{\kappa}+2.35) . \tag{11}$$

The result of this fitting is plotted in Fig. 7(b).

It should be noted that Ditlefsen and Lothe's prediction of Eq. (10) is in disagreement with the original paper of MacDonald and Sarginson[12] who predicted that the value of β at the maximum should decrease as κ decreases. This gives rise to smaller $\omega\tau$(max) than those of Eq. (10). Our findings thus support qualitatively the predictions of MacDonald and Sarginson.

The negative slope of $\Delta R_S(B)/R_S(0)$ for one sample 3 (Fig. 3) is similar to the prediction from the free-electron model by MacDonald and Sarginson[12] and Ditlefsen and Lothe[17] that is shown schematically in Fig. 1. For a thicker sample 5, there is a tendency to a more positive magnetoresistance at high fields. This increase may be from inherent bulk potassium which exhibits a linear magnetoresistance. However, the full cause of this positive Kohler slope is not known because it is even larger than for the bulk samples with a similar purity, studied by Taub *et al.*[14]

The last result of our study is presented in Fig. 6, where we show the data taken with the field oriented

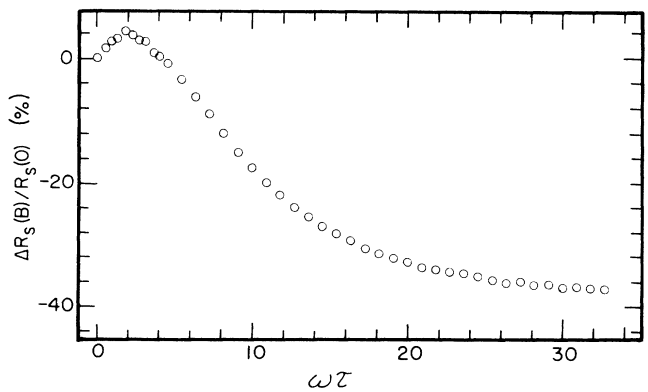

FIG. 3. The transverse magnetoresistance of sample 3 in the MacDonald geometry.

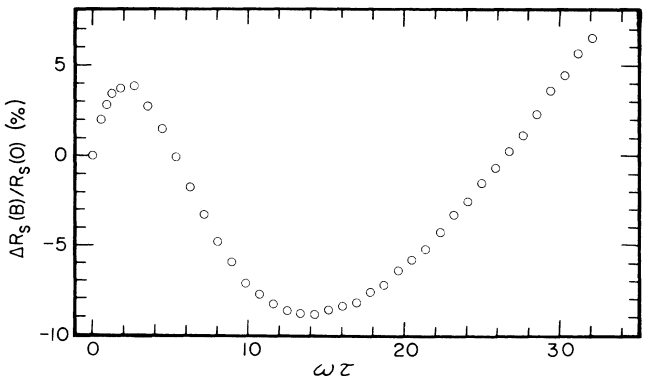

FIG. 5. The magnetoresistance data of sample 3, taken at $\theta=75°$, which shows a clearly resolved minimum, similar to that at $\theta=90°$ for sample 5 shown in Fig. 4.

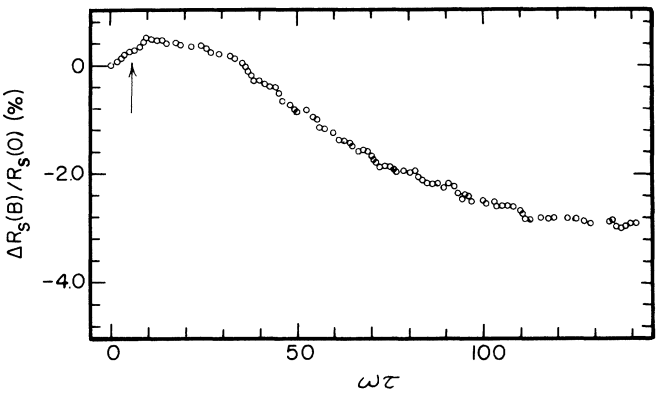

FIG. 6. The longitudinal magnetoresistance of sample 2 with B parallel to the film plane (the angle θ between **B** and **n** is 90°) and to the current I flowing along the longest sample axis L. The arrow indicates the position of the maximum expected from the theoretical prediction of Ref. 23 [see Eq. (12) in text].

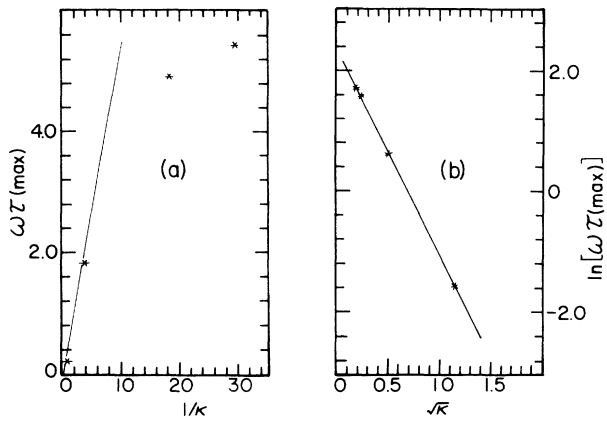

FIG. 7. (a) The position of $\omega\tau$(max) at the maximum in $\Delta R_S(B)/R_S(0)$ for samples 2, 3, 4, and 5 vs $1/\kappa$; the solid line is the theoretical prediction given by Eq. (10). (b) The plot of $\ln[\omega\tau(\max)]$ vs $\sqrt{k}$ to show the fitting of the $\omega\tau$(max) data, by the empirical Eq. (11), which are shown in (a) by a solid line.

parallel to the current but still lying in the film plane (longitudinal magnetoresistance). The theory (MacDonald,[22] Chambers[6]) predicts no initial maximum in the $\Delta R_S(H)/R_B(0)$ for the case of wires. The available theoretical investigation of this geometry in the case of thin metallic films, which was done by Kao,[23] shows that the longitudinal magnetoresistance might give the initial maximum before going to the saturation of the size effects for high fields. Kao gave a rough estimation of the position of this maximum as

$$\beta=d/r=1.26\kappa^{0.57} \quad \text{or} \quad \omega\tau=1.26\kappa^{-0.43}\ , \tag{12}$$

which holds up to 10%. We found good qualitative agreement with the predictions of Kao for this geometry (not yet reported for alkali metals), but the apparent position of the maximum is for a higher value of $\omega\tau$ than the one given by Eq. (12). It is interesting to note that Azbel'[8] estimated on a purely qualitative ground that this maximum should occur at $\omega\tau\sim\kappa^{-0.5}$, which is quite close to the result derived by Kao[23] for this case.

V. CONCLUSION

The magnetoresistance of rolled potassium films was investigated with a sample temperature of 4.2 K and the magnetic field parallel to the sample surface. The film thickness was between 1.8 and 108 μm. The results are reproducible for films prepared without oil. In the MacDonald geometry with the magnetic field perpendicular to the current, there is a resistance maximum for $1<\omega\tau<5$ which depends on film thickness. The $\omega\tau$ at the maximum does not agree with the predictions of Ditlefsen and Lothe from free-electron theory. An empirical relation between this $\omega\tau$ and $\kappa=d/l$ is presented. The magnetoresistance at high fields up to $\omega\tau=32$ depends on sample thickness and it is only qualitatively similar to the predictions of free-electron theory, and only for an intermediate thickness of 20 μm. For thinner samples, the resistance decreases with increasing field up to $\omega\tau=32$. For thicker samples there is a positive Kohler slope in magnetoresistance. It may be from the bulk magnetoresistance of potassium. Size-effect theory based on the free-electron approximation does not explain all the properties of the magnetoresistance in the MacDonald geometry.

The magnetoresistance in the longitudinal geometry has a broad maximum and tends towards saturation at high fields. This does not agree with the predictions of Chambers and MacDonald for wires but does follow qualitatively the theory of Kao.

Since no quantitative agreement was found between our reproducible observations and the available free-electron models of size effects, the development of a new theory is encouraged for the classical size effect of the magnetoresistance in alkali metals.

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