Strain-Dependent Magnetoresistance of Potassium*

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The transverse magnetoresistance of single-crystal potassium has been measured by the helicon method, a probeless technique. The magnetoresistance was found to vary linearly with field in the region $\omega_c\tau\gg1$. This result is in substantial agreement with previous dc measurements. Standard theory cannot explain a linear magnetoresistance. This inability is especially interesting because potassium is thought to be the simplest metal. It was found that Kohler's rule was not obeyed and that strain was an important parameter in the deviation from Kohler's rule. Carefully prepared single crystals of potassium had 40% of the magnetoresistance observed in polycrystalline samples. Cold working increased the magnetoresistance by as much as a factor of 5, and annealing reduced the magnetoresistance. In low-strain single crystals the magnetoresistance was anisotropic; the smallest values were observed for the field parallel to [110] and [123]. The field dependence of the Hall coefficient was also measured. The Hall coefficient of potassium decreased by several percent in the region $\omega_c\tau\gg1$ where standard theory predicts field independence. New theoretical investigations of high-field transport properties and the Fermi surface of potassium will be necessary to explain these observations.

I. INTRODUCTION

We wish to report experimental measurements of the transverse magnetoresistance of single-crystal potassium. The bulk magnetoresistance was determined from helicon-standing-wave resonances,[1] a method not requiring the attachment of probes to the metal. The measurements were performed in the high-magnetic-field regime $\omega_c\tau\gg1$, where ω_c is the cyclotron frequency and τ the relaxation time at zero field. This study was undertaken because previous measurements[2–5] on polycrystalline alkali metals have indicated a linear field dependence of the transverse magnetoresistance; this is in conflict with the theoretical prediction[6] that the transverse magnetoresistance should be independent of magnetic field for $\omega_c\tau\gg1$. Since the alkali metals, especially potassium, are known to have simple electronic structures, and since the general theory of magnetoresistance has been remarkably successful in explaining the observations in more complicated metals,[7] this contrast between experiment and prediction is especially puzzling. This disagreement has often been attributed to experimental uncertainties associated with probe effects and the partial martensitic phase transformation[8] present in lithium and sodium. In the present experiment, the helicon method avoids probe difficulties and the choice of potassium eliminates the complications due to more than one crystalline phase.

Our investigation, using the helicon method, shows the transverse magnetoresistance of single-crystal potassium to have a linear field dependence well into the high-field regime. In addition, qualitative measurements show the magnitude of the linear magnetoresistance to depend on the amount of strain present in the samples under investigation.

II. THEORY OF MAGNETORESISTANCE

A highly successful theory of magnetoresistance has been developed by Lifshitz, Azbel, and Kaganov (LAK).[6] Their semiclassical analysis treats the effect of the magnetic field as a perturbation on the electronic structure determined from band theory. The treatment can be applied to any shape Fermi surface and the only assumption made about the collision integral is that it is independent of magnetic field. The results of the LAK theory are expressed in terms of the resistivity tensor $\rho_{ij}(B)$ which is assumed to relate the current density J_j and the electric field E_i:

$$E_i=\sum_j \rho_{ij}J_j, \quad i, j=x, y, z. \tag{1}$$

If the applied magnetic field is in the z direction, the elements ρ_{xx} and ρ_{yy} are the transverse magnetoresistivity while the element ρ_{zz} is the longitudinal magnetoresistivity. The elements ρ_{xy} and ρ_{yx} are the Hall resistivity. This paper will concentrate exclusively on the transverse magnetoresistivity hereafter referred to simply as the magnetoresistivity.

The general methods and results of the LAK theory are well known.[7] The field dependence of the magnetoresistivity tensor elements is related to the topology of

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the Fermi surface and the relative number of electrons and holes. The important topological factor in the LAK theory is the type of orbit, open or closed, which the carriers describe in reciprocal space in the presence of a magnetic field. The LAK theory has been remarkably successful in explaining the majority of magnetoresistance observations even in metals with complicated Fermi surfaces.[7]

The fact that potassium is an uncompensated metal with electron-like carriers limits the type of resistivity tensors to be expected. For an uncompensated metal, the LAK theory predicts two possible tensors whose forms depend on the presence or absence of open orbits. In metals with closed orbits, the off-diagonal elements should vary linearly with field in the high-field limit, i.e., $\rho_{xy} \propto RB$, where R is the Hall coefficient. The transverse magnetoresistivity ρ_{xx} and ρ_{yy} should become independent of field for $\omega_c\tau \gg 1$. The presence of an open orbit perpendicular to the applied magnetic field has a dramatic effect on the field dependence of the resistivity tensor. If the open orbit direction is parallel to the x axis and the field along the z axis, LAK predict that ρ_{xx} is proportional to B^2. Hence, for single-crystal uncompensated metals, the LAK theory predicts that the transverse magnetoresistivity should either be independent of magnetic field or show a quadratic field dependence. No other field dependence is expected, although it has been suggested that *polycrystalline* metals in which open orbits are present can exhibit a linear magnetoresistance as the result of the average of open-orbit B^2 and closed-orbit B^0 terms. The linear dependence observed in polycrystalline copper has been attributed to this effect.[9,10]

In order to predict the magnetoresistance to be expected in potassium and the other alkalis, it is necessary to know their Fermi surfaces. Extensive experimental work on the alkalis indicates that their Fermi surfaces are all nearly spherical. The most comprehensive experimental results are due to Shoenberg and Stiles[11] whose de Haas–van Alphen measurements on potassium showed the Fermi surface to be spherical to one part in 10^3. Other alkali metals also have been shown to have nearly spherical Fermi surfaces,[11–14] although the experimental evidence is not so clear for sodium and lithium because of their partial martensitic phase change at low temperatures. For those portions of sodium that remain in the bcc phase, the evidence is that the Fermi surface is nearly spherical.[15] This experimental evidence is in good agreement with recent band-structure calculations.[16–18] However, Overhauser[19,20] has suggested that exchange effects could lead to the existence of charge-density waves (CDW) in the alkalis. In the case of potassium this would lead to a Fermi surface which under certain circumstances could support hole-like orbits.[21] Although the question of the CDW in potassium is unresolved at this time, its existence cannot be ruled out on the basis of existing data.[22–24]

If the Fermi surface of potassium is assumed to be spherical, the predictions of the LAK theory are very precise. The transverse magnetoresistance should be independent of field for all fields such that $\omega_c\tau \gg 1$. In addition, the Hall coefficient R should be field independent and equal to $(ne)^{-1}$, where n is the number density of carriers and e is the charge of the electron. It should be stressed that these predictions do not depend on the details of either the collision mechanism or the Fermi surface. All these predictions explicitly assume that magnetic breakdown is not taking place.

In discussing the general magnetoresistance problem, it is necessary to note that the resistance $\rho(B)$ at any given magnetic field depends not only on the magnitude of the magnetic field but also on the resistance at zero magnetic field $\rho(0)$.[25] According to Kohler's rule,

$$\delta \equiv \frac{\rho(B)-\rho(0)}{\rho(0)} = g[B/\rho(0)], \quad (2)$$

where g is a function whose form is independent of B and $\rho(0)$. This relationship is found to be accurate in polycrystalline indium.[26] The LAK theory is consistent with Kohler's rule and gives the form of the function g for various Fermi surfaces.

III. PREVIOUS EXPERIMENTAL MEASUREMENTS OF ALKALI METAL MAGNETORESISTANCE

The original work on the magnetoresistance of the alkali metals was performed by Kapitza,[27] but the first extensive investigation was done by Justi and coworkers.[2] Justi's experimental arrangement was the standard dc, four-probe method. His early work in sodium showed that the resistance increased with magnetic field and that it was anisotropic in a single-

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crystal sample.[28] Since this work was done above 14°K, the measurements were not in the high-field regime, although later work by Justi attained values of $\omega_c\tau$ as high as 10 for sodium and 3 for potassium. The field dependence of the resistance for both metals was approximately linear although a few samples indicated a faster than linear dependence. The important fact was that the resistivity increased with field as much as 35% in sodium and 8% in potassium and did not show signs of saturation.

The analysis of magnetoresistance in terms of the "reduced Kohler variable" $B/\rho(0)$ takes on a particularly simple form if the field dependence is linear:

$$\frac{\rho(B)-\rho(0)}{\rho(0)}=S|\omega_c\tau|\,,\quad \omega_c\tau=RB/\rho(0) \qquad (3)$$

where we shall call the factor S the "Kohler slope." Kohler's rule states that S should be independent of $\rho(0)$ and B, so that the resistance of any sample, given $\rho(0)$ and B, should be specified by S. In contrast to the LAK theory, which predicts that S should tend to zero at large fields, Justi found S values in the neighborhood of 3×10^{-2} for both sodium and potassium.

MacDonald[3] investigated the magnetoresistance of sodium and rubidium using the dc probe method and extended $\omega_c\tau$ to 50 or more. He concluded that the linear increase in the magnetoresistance persisted well into the high-field regime and that there was a dramatic anisotropy of the magnetoresistance in polycrystalline samples. His values of S were as low as 4×10^{-2} and as high as 10^{-1} for sodium. S values as high as 1.5×10^{-1} were observed for rubidium. MacDonald noted that probe effects could have influenced his measurements since current paths near the probes would be very field-dependent. For certain sample geometries, MacDonald observed a resistivity that decreased with magnetic field in sodium. This result was almost certainly due to a mixing of Hall voltage with loss voltage caused by probes. Bowers[4] and Rose[5] repeated the probe measurements of MacDonald on sodium and obtained similar results.

The first detailed study of the magnetoresistance of the alkalis by probeless methods was carried out by Rose using the helicon-wave techniques. His samples were polycrystalline and showed the same linear dependence exhibited in the probe measurements up to the highest attainable $\omega_c\tau$ values of 150 in sodium and 70 in potassium, although the value of S was less than that of probe methods for both metals. The result of the helicon measurement was most dramatic for sodium where he found a slope of 2×10^{-3} compared with MacDonald's result of 2×10^{-2}. In potassium, Rose found a slope of 1×10^{-2} by helicon measurements as compared to the 2×10^{-2} reported by Justi. Helicon measurements on lithium gave slopes of 15×10^{-2} or more, comparable to the results of probe methods.

[28] Whether the sodium sample was indeed a single crystal is unclear owing to the martensitic phase transformation.

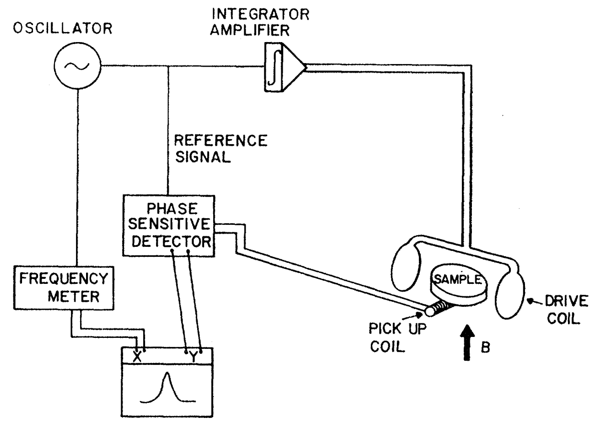

FIG. 1. Experimental configuration and electronic schematic. The disk-shape sample was oriented perpendicular to the static magnetic field B (z axis). An exciting magnetic field was produced by a Helmholtz pair (x axis). The helicon was detected by a coil underneath the sample (y axis). The sample could be constrained with a force applied to the top of the sample. The electronic components were standard audio-frequency devices.

The most important conclusion of Rose's work was that the helicon gave the same qualitative magnetoresistance measurements as probe methods, i.e., a linear dependence. The Kohler slope seemed to be less for sodium and potassium when measured by a probeless technique. The conclusion was that probe effects were important, but were not the sole cause of the linear magnetoresistance of the alkalis.

Rose also improved the probe method of measuring the magnetoresistance of potassium.[5] A significant problem in obtaining a reliable measure of the voltage due to the resistance has always been the presence of a large Hall voltage. Rose used a sample geometry in which the loss voltage was much larger than the Hall voltage. The sample was wound in a helix, the axis of the helix being parallel to the magnetic field. The transverse magnetoresistance (the pitch of the helix was small) was obtained from a voltage measured over the total length of the wire while the Hall voltage was generated over the diameter of the wire only. In this more reliable geometry, Rose found the magnetoresistance increased with field linearly, S values being on the order of 10^{-2} for sodium, potassium, and lithium.

IV. HELICON METHOD FOR MEASUREMENT OF MAGNETORESISTANCE AND HALL COEFFICIENT

Information about the magnetoresistivity tensor [Eq. (1)] can be deduced from helicon-standing-wave resonances. Chambers and Jones were the first to develop this experiment.[1] Our method was very similar, extending their work to high magnetic fields and single crystals. The coil system used in this experiment is shown in Fig. 1. The disk-shaped sample was placed

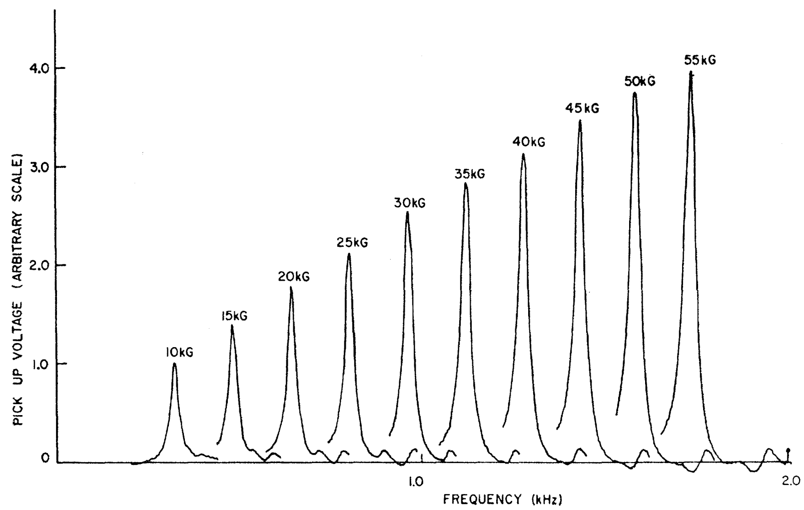

FIG. 2. Typical helicon resonances in potassium. The pickup voltage in phase with the drive current was plotted on an xy recorder as a function of frequency for different magnetic fields. The sample was a single crystal of potassium, 8 mm in diameter and 1.4 mm thick; the magnetic field was parallel to the [111] direction; the RRR was 3000.

between a pair of pseudo-Helmoltz coils. These coils produced a linearly polarized magnetic field in the x direction. The sample plane was defined as the x-y plane and the large, static magnetic field was placed along the z direction. The pickup coil, its axis along the y direction, detected the circularly polarized helicon wave. Since we wished to exert a uniaxial stress along the z direction, the pickup coil was placed underneath the sample, instead of enclosing the sample; the resulting loss of signal did not present a problem.

The response of such a coil system has been derived by Chambers and Jones[1] and extended to anisotropic magnetoresistivity tensors by Penz.[29] Using the formalism of Eq. (1), the pickup voltage will be proportional to the transverse permeability μ_T:

$$\mu_T=\frac{8}{\pi^2}\frac{\rho_{xy}}{\rho_{xx}+\rho_{yy}}\sum_{m=1,\text{odd}}^{\infty}\frac{1}{m^2}\frac{1}{1+iQ(\omega/\omega_m+\omega_m/\omega)},$$

where

$$\omega_m=\frac{\pi^2m^2}{\mu_0d^2}(\rho_{xx}\rho_{yy}-\rho_{xy}\rho_{yx})^{1/2}\underset{\text{isotropic case}}{\longrightarrow}\frac{\pi^2m^2}{\mu_0d^2}(R^2B^2+\rho^2)^{1/2},$$

$$Q=\frac{(\rho_{xx}\rho_{yy}-\rho_{xy}\rho_{yx})^{1/2}}{\rho_{xx}+\rho_{yy}}\underset{\text{isotropic case}}{\longrightarrow}\frac{(\rho^2+R^2B^2)^{1/2}}{2\rho},$$

and d is the sample thickness. The implications of these equations have been discussed elsewhere. It is necessary to emphasize that the helicon measures the sum of the resistances in the xy plane: $\rho_{xx}+\rho_{yy}$. This is a well-defined average and enables the confident analysis of single-crystal data. The resonant frequency is proportional to $(-\rho_{xy}\rho_{yx})^{1/2}$ when $\rho_{xy}\gg\rho_{xx}$, ρ_{yy}. In the isotropic case, $\omega_m\propto RB$ for $RB/\rho\gg1$.

The alternating fields in the drive coil acted with the static magnetic field to exert torques on the coil system. Two sets of phosphor bronze springs, one above and one below the sample, held the coil system in the magnet and severely constrained this movement.

The static magnetic field was produced by a superconducting solenoid which had a $\frac{15}{16}$ inch bore, a maximum magnetic field of 55 kG and a uniformity of 1% over a 1-in. sphere. Work was also performed in high-field Bitter solenoids at the National Magnet Laboratory (NML), Cambridge, Mass.

The electronic circuit used for detection of the helicon resonance is shown in Fig. 1.[30] The components of the system were standard low-frequency units, the maximum frequencies being approximately 5 kHz. The active element of the phase-sensitive detector was a semiconductor Hall-effect multiplier. Since the signal-to-noise ratios of the signals across the pickup coil were on the order of 10, phase-sensitive detection was a convenience rather than a necessity. It also allowed the measurement of the real and imaginary parts of μ_T which are simpler to analyze than the modulus of μ_T. Care was taken to assure that the reference signal was either in phase (real part of μ_T) or out of phase (imaginary part of μ_T) with the drive current.

A tracing of experimental data for a plate of potassium 1.4 mm thick and 8 mm in diameter is shown in Fig. 2. The output of the phase-sensitive detector is plotted versus frequency for 10 to 55 kG in 5-kG steps. All resonances shown are the $m=1$ or fundamental resonances. The factors to be noted are the large Q's, the good signal-to-noise ratios, and the low fundamental frequencies.

A helicon propagating in a metal is equivalent to a rotating magnetic moment. The interaction of the static magnetic field with this magnetic moment produces a torque which tends to twist the moment out of the plane of rotation. If the sample is not constrained externally the resulting sample motion will remove energy from the helicon wave and in general degrade the resonance. This effect is so large that at 50 kG it is

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nearly impossible to observe a resonance in a frees standing potassium disk.

Previous experimenters have constrained samples with frozen mineral oil. Because of the large difference in contraction rates, however, this method exerts large stresses on the sample, and for soft materials such as potassium there is a large resultant strain. Since strain could play a role in the anomalous magnetoresistance, it was necessary to devise a method to hold the sample under small stress. Compressional stress could be exerted on the sample by a plunger which was connected by a stainless steel tube to the room-temperature environment. A known stress could be applied to the sample by adding weights to the top of the tube.

A great deal of work was invested in the support problem since the effect of unwanted sample motion is to cause a decrease in the Q of the helicon resonance, a degradation in resonance shape, and an apparent breakdown of the helicon dispersion relation. These effects are field-dependent and tend to enhance the measured magnetoresistance. The linear magnetoresistance observed in potassium, however, persisted even when the sample was rigidly cemented in frozen mineral oil; it is difficult, therefore, to attribute this effect to unwanted sample movement.

Measurements were taken using the minimum stress necessary to achieve a signal which was stress-independent at 50 kG. This method of sample support is defined as the "minimum stress condition." The stress necessary to produce reliable helicon signals was about 4–8 kg/cm^2; this compares with the yield stress of potassium at 4.2°K of 20 kg/cm^2.[31] Since the samples were not constrained until they were in liquid helium, the support stress was in the region of elastic strain. Measurements were also performed using the frozen-mineral-oil technique.

The sample resistance can be calculated from a helicon-resonance curve in two ways: the helicon amplitude is proportional to $RB/\rho(B)$ for $\omega_c\tau\gg1$ and thus can yield the relative magnetoresistance directly. Secondly, the Q of the helicon resonance is also proportional to $RB/\rho(B)$ and, although more difficult to obtain experimentally than the amplitude, is a convenient way to confirm the results of the amplitude measurement. Values of the magnetoresistance of potassium obtained by both methods agreed closely in this experiment, although the first method was used predominately, since it allowed a more rapid accumulation of data.

It is necessary to describe the extent to which the experiment conformed to the boundary conditions of the theory. The theory assumed an infinite plate, and the samples used were obviously not infinite in extent. As a result a series of minor mode resonances corresponding to solutions of the three-dimensional boundary-value problem were observed.[32] Although the three-dimensional boundary-value problem has not been solved analytically, Chambers and Jones have shown that the fundamental helicon resonance in finite samples should have the same shape and magnitude as in an infinite plate. Once the fundamental has been distinguished from the minor modes, the infinite plate theory should be applicable to the finite plate experiments. In practice, distinguishing the fundamental mode from the minor modes presents no problem since the amplitudes of the latter are greatly attenuated. The disks used in the present experiment had diameters of approximately 8 mm and thicknesses near 1 mm; the minor modes were usually separated from the fundamental for all fields above 10 kG.

Additional complications can arise from sample imperfections. Surface roughness, internal inhomogeneities, and nonparallel crystal faces can distort the helicon wave in a more or less unpredictable way. In order to determine the extent of these effects, we performed experiments on intentionally deformed samples; our conclusion is that none of the imperfections encountered in our samples could account for the anomalous linear magnetoresistance seen in potassium.

Abundant evidence exists which indicates that the helicon gives a reliable method for measuring magnetoresistance and Hall coefficients to within accuracy of a few percent. Measurements of the magnetoresistance of the noble metals by the helicon give results similar to probe measurements.[33,34] The mere observation of the helicon in compensated metals for certain orientations confirms that the helicon measures the Hall resistivity ρ_{12} correctly.[33] Measurements of the magnitude of the Hall coefficient by the helicon method agree to within a few percent with theory.[1]

In an effort to calibrate our experimental system, we have measured the magnetoresistance of Al and In. For polycrystalline In we found that the magnetoresistance was linear or almost linear in the regime $\omega_c\tau\gg1$. This result was also seen by Gaidukov[35] for conventional probe measurements. Our method is not designed to measure the region $\omega_c\tau\sim1$ where a definite knee appears in the magnetoresistance of indium.[36] Our results in Al were similar to probe measurements.[37,38]

The measurement of the potassium zero-field resistance $\rho(0)$ in these experiments was accomplished by extrapolating the helicon data to zero field, and by the use of the inductive method developed by Bean *et al.*[39]

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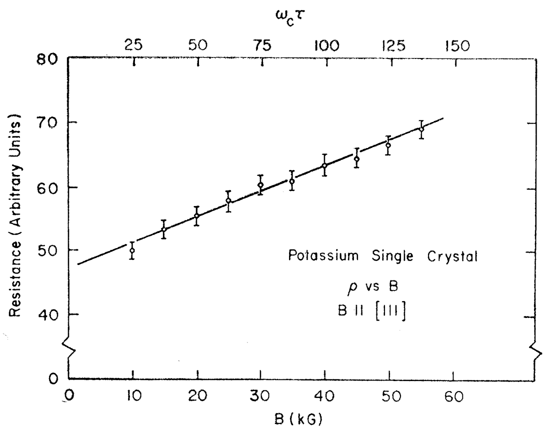

FIG. 3. The magnetoresistance of single-crystal potassium. The magnetoresistance was deduced from the helicon resonances shown in Fig. 2. The resistance increases linearly with magnetic field; at $\omega_c\tau=140$ it has increased 40% over its zero-field value.

The two methods gave results which agreed to within the experimental error of $\pm5\%$ although the former method was preferred since it did not require additional handling of the sample. The helicon data were extrapolated to zero from measurements at $\omega_c\tau>20$; in this regime the minor modes were well separated from the fundamental.

V. SAMPLE PREPARATION

The potassium used for these experiments was purchased from Mine Safety Appliances (MSA), Callery, Pa.; the purity of the stock was specified as 99.95%. The residual resistance ratio (RRR) of this material at 4.2°K was approximately 3000.

Single crystals were grown in a Bridgman-type stainless steel crucible filled with xylene.[40,41] The xylene prevented the molten potassium from wetting the crucible and protected the highly reactive metal. Since the potassium contracted upon solidification, it was possible to slide the charge from the crucible without straining the crystal. The purity of the metal was not noticeably affected by this procedure. The single crystals were etched in a 1% mixture of absolute ethyl alcohol and xylene in order to achieve sharp crystallographic etch planes. These planes were used to orient the crystal by an optical technique developed by Grimes[13,42] which had an alignment accuracy of $\pm2°$.

Single crystal disks with thicknesses of approximately 1 mm and elliptical surfaces with 4-mm semiminor axes were cut with a string saw. Although previous experimenters have lapped their crystals to achieve smooth, parallel faces, it was found that this resulted in polycrystalline samples for plates as thin as 1 mm. Every effort, therefore, was made to cut parallel faces directly with the string saw. It was, however, impossible to avoid small grooves in the faces of the crystals. Most of the samples produced in this manner, however, were sufficiently parallel and smooth to produce sharp, well-defined helicon resonances. After the samples were cut, they were stored in mineral oil and annealed at room temperature for 24 h.

Polycrystalline samples were fabricated by pressing the potassium stock between stainless steel plates. The metal was then etched clean, and the etch revealed four or five crystallites on a sample 8 mm in diameter.

There were two distinct methods of holding the sample, one with the minimum stress apparatus, and the other using frozen mineral oil. When frozen mineral oil was used, the sample was etched clean and then immersed in mineral oil. The mineral-oil-coated sample was immediately placed in the coil system and extra oil was poured over the sample and coil system. The coil was then quenched in liquid nitrogen.

For the minimum stress configuration, the sample was first etched clean, and the xylene blotted off. Samples were immediately quenched in liquid nitrogen and stored until ready for use. The samples were loaded into the coil system at temperatures near 77°K and then

TABLE I. The magnetoresistance of single-crystal potassium.[a] $S_{av}=0.31\times10^{-2}$; $\Delta_s=0.15\times10^{-2}$=standard deviation; $\Delta_s/\sqrt{N}=0.03\times10^{-2}$.

	$[hkl]$[b]	$\delta(55)$[c]	$\omega_c\tau(55)$[d]	$10^2 S$[e]	$10^3\times$(RRR)[f]
1	100	45	133	0.34	3.1
2	100	62	146	0.42	3.4
3	100	38	143	0.26	3.4
4	100	30	56	0.54	1.3
5	100	25	46	0.54	1.1
6	100	42	64	0.65	1.5
7	100	27	63	0.43	1.5
8	100	15	61	0.25	1.4
9	110	28	143	0.20	3.4
10	110	7	39	0.18	0.9
11	110	10	54	0.18	1.3
12	110	12	61	0.20	1.4
13	110	19	74	0.26	1.7
14	110	5	50	0.10	1.2
15	110	6	60	0.10	1.4
16	111	90	165	0.55	3.9
17	111	40	127	0.31	3.0
18	111	45	136	0.33	3.2
19	123	51	92	0.55	2.2
20	123	30	139	0.22	3.3
21	123	77	166	0.46	3.9
22	123	13	59	0.22	1.4
23	123	5	49	0.10	1.2
24	123	10	100	0.10	2.4

[a] Crystals are in stress state A, defined as single-crystal samples held under minimum stress condition. All samples were elliptical disks with semiminor axes of 4 mm, semimajor axes that varied from 4 to 6 mm, and thicknesses that ranged from 1 to 1.5 mm.
[b] $[h,k,l]$ represents the Miller crystallographic indices of the magnetic field and the disk normal.
[c] $\delta(55)=[\rho(55)-\rho(0)]/\rho(0)$. $\rho(0)$ determined by linear extrapolation of helicon magnetoresistance data to zero field; 55 denotes 55 kG.
[d] $\omega_c\tau=2Q(55)[\rho(55)/\rho(0)]$.
[e] $S=[\rho(55)-\rho(0)]/2Q(55)\rho(55)$ = Kohler slope.
[f] RRR $=\omega_c\tau(55)/RB\times6.1$ μΩcm = residual resistivity ratio, where B is 55 kG.

[40] W. B. Daniels, Phys. Rev. **119**, 1246 (1960).
[41] P. A. Penz, Ph.D. thesis, Cornell University, 1967 (unpublished).
[42] C. C. Grimes, Ph.D. thesis, University of California (unpublished).

the free-standing sample was slowly lowered into the liquid helium. Thus very little stress was exerted on the sample before an experiment was begun.

VI. EXPERIMENTAL DETERMINATION OF THE MAGNETORESISTANCE OF SINGLE-CRYSTAL POTASSIUM

The resonances from a typical helicon experiment using a single crystal of potassium were given in Fig. 2; the analysis of this data for the magnetoresistance is shown in Fig. 3. The sample normal of the single-crystal disk used in these measurements was aligned along the [111] axis, and the sample was held in place by a uniaxial pressure which was approximately 20% of the yield stress. The anomalous linear magnetoresistance is seen to be quite pronounced for this sample; at 55 kG the resistance is 44% greater than the extrapolated zero-field resistance, corresponding to a Kohler slope S of 0.32×10^{-2}. Since the helicon measures the resistance in the plane perpendicular to the magnetic field, Fig. 3 is a plot of $\rho_{xx}+\rho_{yy}$, where x is parallel to $[12\bar{3}]$ and y is parallel to the $[\bar{5}41]$ direction.

Twenty-four single crystals were measured under minimum stress conditions. The data shown in Fig. 3 is typical of 19 other single crystals whose resistance was found to have a field dependence which was linear to within the +2% estimated error. Four other single crystals exhibited an increase that was principally linear, but the scatter of the data was somewhat larger than that shown in Fig. 3. The results of these measurements are presented in Table I. The linear field dependence is consistent with most of the previous probe measurements on alkali metals, especially the helix geometry probe measurements of Rose[5] which are presently the best available dc measurements on the

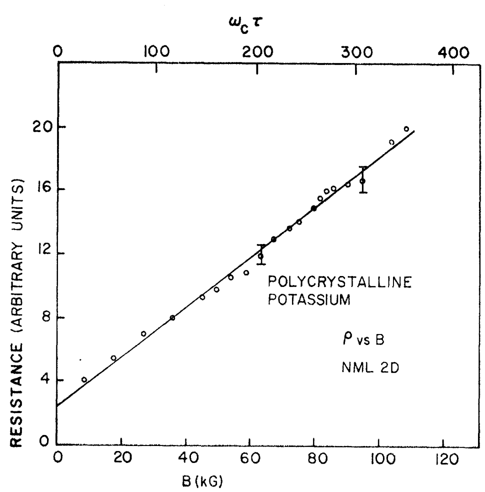

FIG. 4. The magnetoresistance of potassium at very high fields. The magnetoresistance of polycrystalline potassium was observed to increase linearly for $\omega_c\tau$'s approaching 400. The measurements were performed in a conventional solenoid at the Bitter National Magnet Laboratory.

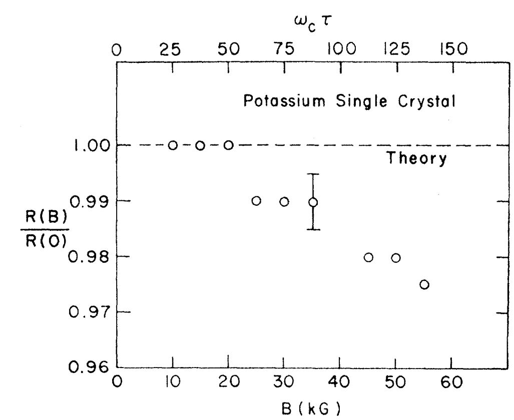

FIG. 5. Field variation of the Hall coefficient in single-crystal potassium. The Hall coefficient relative to its zero-field value was obtained from the helicon resonances shown in Fig. 2. A small decrease in $R(B)/R(0)$ can be detected; the experimental error of $\pm\frac{1}{2}\%$ is shown by a typical error bar.

alkalis. Since the linear magnetoresistance is observed in two entirely different experimental configurations, we conclude that it is a property of single-crystal potassium metal; probe effects or mixed crystallographic phases may enhance the linear term, but are not the prime cause.

We have also measured the magnetoresistance of polycrystalline potassium up to 110 kG in a Bitter solenoid at the NML. Figure 4 shows the resistance of a polycrystalline sample held in the coil system with frozen mineral oil. It is evident that the linear magnetoresistance which we have observed in single crystals is also present in polycrystals and shows no sign of saturating. The uncertainty in the measurements of ±5% is due to the large amount of noise in the Bitter solenoid, and to mechanical resonances which are more troublesome at higher fields. The increase in resistance over the zero-field resistance is greater than a factor of 7 at $\omega_c\tau$ of 350.[43]

The Hall coefficient can also be measured from helicon resonances.[1] While the principal emphasis of this research was on magnetoresistance, work was done on the measurement of the Hall coefficient. Figure 5 shows the field dependence of R as determined from the single-crystal data of Fig. 2. There is a tendency for R to decrease with field, although the change is not large with respect to the experimental uncertainty of $\pm\frac{1}{2}\%$. It was found, however, that the decrease of a few percent at 55 kG was observed generally in the poly- and single-crystal samples tested.

The field dependence of R was also measured up to 110 kG. In Fig. 6, a plot of R versus B is presented for

[43] The authors have previously reported a magnetoresistance in potassium that gave evidence of a quadratic as well as a linear field dependence [P. A. Penz and R. Bowers, Bull. Am. Phys. Soc. 11, 92 (1966)]. Additional analysis of the experiments indicates that the quadratic field dependence probably was an experimental artifact.

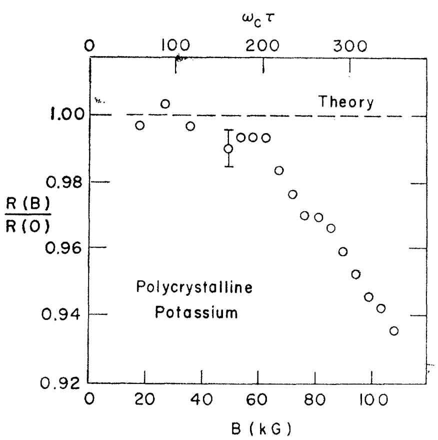

FIG. 6. Field variation of the Hall coefficient at very high fields. The relative Hall coefficient was observed to decrease by over 6% at $\omega_c\tau$ of 350 polycrystalline potassium; the experimental error of $\pm\frac{1}{2}\%$ is shown by a typical error bar.

the same polycrystalline K sample that produced the magnetoresistance data of Fig. 4. The decrease of 6% observed at 110 kG is well outside the experimental error. This decrease is not large, but is significant since the LAK theory predicts a field-independent Hall coefficient. In contrast to potassium, no field dependence was observed in an aluminum sample of comparable $\omega_c\tau$, within the experimental error. It should be remembered that the quantity being measured is $(-\rho_{xy}\rho_{yx})^{1/2}/B$, which we are calling the Hall coefficient. The data could equally well be interpreted as demonstrating a Hall resistivity of the form $\rho_{xy}=B/ne+C_1B^2+C_2B^3\cdots$, i.e., with higher-order coefficients being nonzero. The LAK theory predicts that all higher-order coefficients should be zero.

In order to measure the magnitude of R, it is necessary to measure sample thicknesses accurately. Since we wished to avoid unnecessary handling of our samples, we measured R for only a few crystals. For these R was found to be in the range -47 to -50×10^{-4} m^3/C when $\omega_c\tau\sim10$. These values are consistent with the low-field ($B<7$ kG) measurements of Chambers and Jones.[1] The discrepancy between our measurements and the free electron value for R of -44.5×10^{-11} m^3/C can in part be accounted for by the fact that a finite-size sample was used. Amundsen[44] has shown in indium that the boundary-value problem correction for a finite-size sample will tend to decrease the magnitude of $|R|$ which is computed from an infinite sample dispersion relation. It remains to be demonstrated for potassium that this correction will account for all the discrepancy between theory and experiment.

VII. EFFECT OF STRESS ON MAGNETORESISTANCE

It has been shown that the magnetoresistance of both single crystals and polycrystals of potassium deviates significantly from the LAK theory. Since the field dependence of the resistance in the anomalous region is linear, the Kohler slope S is the natural measure of the extent of this deviation.

Conversations with Professor J. W. Wilkins and Dr. Z. S. Basinski prompted us to consider the effects of sample strain on the magnetoresistance. Unfortunately, the high reactivity of potassium makes it difficult to measure directly the strain and associated defect density. In view of this fact, we decided to first determine the effect of applied stress on the Kohler slope of the magnetoresistance; it is obvious, however, that there is only a qualitative correlation between crystal strain and applied stress.

A rough measure of the amount of stress applied to our potassium crystals can be made by distinguishing between two stress categories—the unavoidable stress involved in sample preparation, and the stress required to hold the samples in place during the experiment. The two methods for preparing samples were the low-stress, acid cutting of single crystals, and the high-stress method for producing polycrystalline plates. The prepared samples were then held in place in either a minimum-stress configuration (Fig. 1) or a high-stress configuration in which the sample was frozen in place with mineral oil.

The total stress state of any sample is then a combination of the preparation and holding stresses. The four possible stress states, using this qualitative description of stress, are summarized in Table II, along with the results of measurements made on samples in all four states. State A is the lowest stress state, where the sample is a single crystal and is held under minimum stress, state B is defined as a single crystal held with frozen oil, and state C is a polycrystalline sample held under minimum stress. The highest stress state is state D in which the sample is polycrystalline and held with frozen oil. Stress state D is the state in which Rose performed his helicon measurements. Stress state A is the state for which the measurements of Table I were taken.

Under each stress state in Table II is listed the average Kohler slope S_{av}, the number of samples N over which the average was taken, the standard deviation of the measurements Δ_S, and the standard deviation of the

TABLE II. Comparison of stress states. (The purity of the samples used for states B, C, and D was similar to that of state A, Table I.)

State	Stress of preparation	Stress of holding	N[a]	10^2 S_{av}[b]	10^2 Δ_s[c]	10^2 $\Delta_s/\sqrt{N}$
A	Single	Minimum[d]	24	0.31	0.15	0.03
B	Single	Oil[e]	13	0.43	0.25	0.07
C	Poly.	Minimum	14	0.72	0.34	0.09
D	Poly.	Oil	15	0.83	0.34	0.09

[a] N = number of samples tested.
[b] S_{av} = average slope of N samples.
[c] Δ_s = standard deviation of N measurements.
[d] Minimum stress conditions defined in Sec. IV.
[e] Oil—sample held with frozen mineral oil.

[44] T. Amundsen, Proc. Phys. Soc. (London) 88, 757 (1966).

TABLE III. Orientation dependence of S. State A: single crystals, minimum stress holding. The symbols are the same as used in Table II.

[*hkl*]	N	$10^2\ S_{av}$	$10^2\ \Delta_s$	$10^2\Delta_s/\sqrt{N}$
100	8	0.43	0.14	0.05
110	7	0.18	0.05	0.02
111	3	0.40	0.10	0.06
123	6	0.28	0.19	0.08

average $\Delta_S/\sqrt{N}$. The main point of interest in Table II is that the average slope varies significantly among the four stress states. Obviously the values of S_{av} correlate well with the stress states, low stress corresponding to a low value of S_{av}. The Kohler slopes for single crystals, states A and B, are much less than the previous measurements of 1×10^{-2} by Rose[5] and 3×10^{-2} by Justi[2] in polycrystalline K.

It is of interest to note the relatively large standard deviation for the slope measurements within each stress category of Table II. In each case, the standard deviation is much larger than the ordinary measurement errors, apparently reflecting the difficulty in controlling precisely the amount of stress even within a given category. Thus factors such as the original strain of the stock material and minor variations in sample preparation and holding can result in widely varying Kohler slopes even for two samples prepared under very similar conditions. As expected, however, the variation in S values is smaller for single-crystal samples than for the less well-controlled polycrystal samples.

Table III summarizes the dependence of the Kohler slope on the crystallographic orientation for the 24 single-crystal samples in stress state A. The crystallographic indices [*h*,*k*,*l*] represent the crystallographic direction of the magnetic field and the normal to the sample. It can be noted from Table III that there is a perceptible anisotropy in the slope between the directions [110], [123] and [111], [100]. All four directions exhibited a linear magnetoresistance, but the slope for the [110] and [123] directions were less on the average.

Even though stress state B involved single crystals, too few crystals of any one orientation were measured to obtain a meaningful average. It should be mentioned, however, that two samples whose normals were parallel to [123] had especially small slopes of 0.03×10^{-2}, among the smallest observed. Within the experimental error of 0.03×10^{-2} these two samples exhibited a field-independent resistance. The (123) and (110) planes are favored slip planes in potassium.[45]

VIII. EFFECT OF STRAIN ON MAGNETORESISTANCE

In order to investigate further the effect of stress on the magnetoresistance of potassium, we measured the magnetoresistance of individual samples subjected to various controlled stresses.

[45] E. N. Andrade, Proc. Phys. Soc. (London) **52**, 1 (1940).

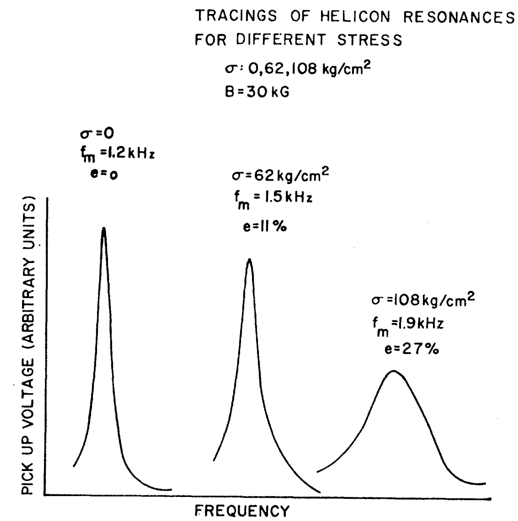

FIG. 7. The influence of applied stress on helicon resonances. The figure is a tracing of three helicon resonances for applied stresses of 0, 62, and 108 kg/cm². The stresses were applied at 77°K. The resonances were all observed at 30 kG, and the increase in resonant frequency f_m indicates a decrease in sample thickness. The decrease in Q and height of the resonances indicates that the resistivity is increasing with stress.

The experimental arrangement for exerting stress on the sample was similar to that in Fig. 1; a compressional stress of up to 100 kg/cm² could be applied to the sample by means of a spring-loaded plunger. Since the applied stresses tended to decrease the thickness of the sample, the plunger mechanism was carefully aligned so that the sample was always pressed between parallel surfaces. The stress was first applied in liquid nitrogen, and the sample was then immediately transferred into liquid helium; in this way annealing and surface deterioration were held to a minimum. Once at 4°K, the pressure was lowered so that the measurements were performed under a holding stress about equal to the yield stress of 20 kg/cm²; this stress was considered inconsequential compared to the stress at 77°K which yielded plastic deformation. Once a set of helicon resonances was traced, the sample was returned to liquid nitrogen and the whole process was repeated at a higher stress.

It was possible to measure the sample strain produced by the process because the applied stress tended to decrease the sample thickness and increase the frequency of the helicon resonance. If f_σ and f_0 are the resonance frequencies at stress σ and zero, their ratio is

$$f_\sigma/f_0=(d_0/d_\sigma)^2\,,\quad (B=\text{const})$$

assuming a strain-independent Hall coefficient—a valid assumption, since all measurements were performed at nearly the same holding stress, and since previous experiments have shown that the Hall coefficient is not sensitive to stress in this range.[46] Here zero stress represents the fact that stress was not applied at 77°K.

[46] T. Deutsch, W. Paul, and H. Brooks, Phys. Rev. **124**, 753 (1961).

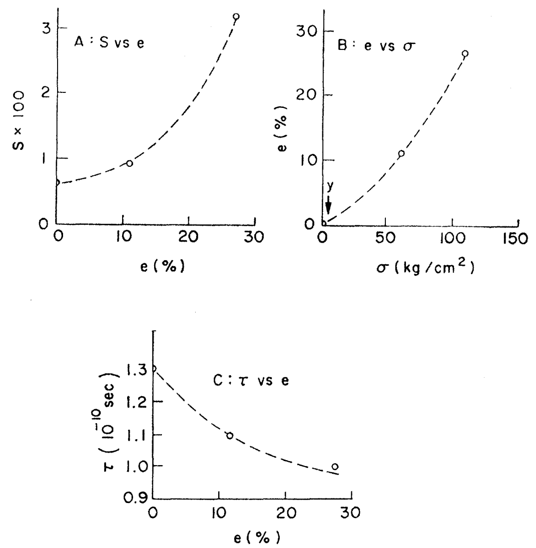

FIG. 8. The influence of strain on the magnetoresistance of potassium. (A) Plot of the Kohler slope S as a function of strain, e. S increases monotonically with strain. (B) Plot of strain as a result of applied stress σ; Y is the yield stress for potassium at 77°K. (C) Shows that the relaxation time τ decreases as a function of strain, presumably as a result of the introduction of lattice imperfections.

The fractional strain e is defined as

$$e \equiv (d_0 - d_\sigma)/d_\sigma = \sqrt{(f_\sigma/f_0)} - 1.$$

The results of stressing a single crystal of potassium are shown in Fig. 7. Three helicon resonances at 30 kG are traced for a sample which was stressed at 77°K with 0, 62, and 108 kg/cm². The effect of increasing applied stress was to broaden and attenuate the resonance and increase its frequency f_m. The magnetoresistance continued to be linearly dependent on magnetic field.

The observed strain dependence of the Kohler slope is shown in Fig. 8 A. S can be seen to increase monotonically with strain. Figure 8 B shows the relationship between crystal strain and compressional stress at 77°K. The arrow labeled y indicates the tensile yield stress of potassium.[31] Plastic deformation was detected experimentally since the resonant frequency of the sample increased each time after the stress had been applied and removed. Although the sample was a single crystal at the beginning of the experiment, the plastic deformation produced a polycrystalline mosaic, whose average crystallite size was several times smaller than those produced by deformation at room temperature.

Figure 8 C presents some approximate data on the effect of plastic deformation on the relaxation time as determined by the resonance Q's. The relaxation time was observed to decrease with strain. The sample was sufficiently pure such that τ at 4.2°K was principally determined by impurity and defect scattering. Plastic deformation is well known to increase the number of defects, and this presumably was responsible for the decrease in τ. We estimate that an applied stress of 100 kg/cm² produces a dislocation density in potassium of about 4×10^{10} cm^{-2}.[47]

Measurements were made on a variety of samples, and the results were similar to those shown in Fig. 8. One test involved an annealing study. After a series of stress measurements were made on one sample, during which its S increased by 100% at a strain of 32%, it was removed from the helium bath and etched in a mixture of xylene and alcohol. Inspection revealed the crystal to have a mosaic structure consisting of 10–15 small crystallites. The sample was then annealed for 4 h in mineral oil at room temperature and returned to the helium bath. Subsequent measurements showed the Kohler slope and the residual resistance ratio of the sample to be restored to almost its original value. This evidence supports our belief that strain is a relevant parameter in the linear magnetoresistance of potassium.

IX. CONJECTURE ON THE ANOMALOUS MAGNETORESISTANCE OF POTASSIUM

The observed magnetoresistance of single-crystal potassium is not explained by the semiclassical theory of an infinite, perfect lattice (LAK). Our experiment only approximates the theoretical conditions: The sample is finite in size and imperfect in structure, and the possibility of magnetic breakdown is present.

Azbel[48] has solved the Boltzmann equation for a bounded metal in the presence of a magnetic field. He finds that boundary scattering can have an important effect on galvanomagnetic theory. He predicts a linear magnetoresistance for uncompensated, closed-orbit metals due to a static skin effect. For the effect to be important, however, the magnetic field must be parallel to the surface and the scale of the surface roughness must be small with respect to the size of typical cyclotron orbits. Neither of these conditions is met in the present experiment, and so it must be concluded that the static skin effect is not responsible for the anomalous results in potassium.

Herring predicts[49] a linear magnetoresistance due to isotropic, bulk inhomogeneities. Physically, the extra loss mechanism results from currents being forced by inhomogeneities to travel parallel to Hall fields. In order to evaluate the relevance of Herring's theory to this experiment, it is necessary to calculate the amount of spatial variation which would be necessary to produce the observed Kohler slope. For the case of an isotropically varying density of carriers n, Herring predicts

$$S = \frac{\langle (n - \langle n \rangle)^2 \rangle}{\langle n \rangle^2},$$

where $\langle\ \rangle$ stands for a spatial average. To produce a typical S of 10^{-2} requires a variation of 10% in the

[47] T. E. Mitchell, *Progress in Applied Materials Research* (Temple Press, London, 1964), Vol. 6, p. 117.
[48] M. Azbel, Zh. Eksperim. i Teor. Fiz. 44, 983 (1963) [English transl.: Soviet Phys.—JETP 17, 667 (1963)].
[49] C. Herring, J. Appl. Phys. 31, 1939 (1960).

carrier density over the sample. This is an unreasonably large variation of n for a metal. Consequently the quantitative fit of Herring's theory to the experiment does not appear promising. It is clear, however, that the correlation of the linear magnetoresistance with strain suggests that further theoretical investigation of the effects of defect structure on magnetoresistance is necessary.

Magnetic breakdown could also cause a departure from the semiclassical LAK theory.[50] This process has not been considered important in potassium because the free-electron sphere does not intersect the first Brillouin zone, i.e., it is a single-sheet Fermi surface. If a mechanism existed that could convert the free-electron sphere into several sheets, magnetic breakdown would certainly cause a departure from a saturation behavior. Overhauser's charge-density wave should be considered in this connection because of its potential for distorting the free-electron sphere.[21]

X. CONCLUSIONS

The transverse magnetoresistance of potassium has been studied using the helicon-standing-wave technique. We have observed a linear field dependence of the magnetoresistance in a wide variety of single-crystal and polycrystalline samples at fields for which $\omega_c\tau \gg 1$. These results are consistent with the best available probe measurements and represent a striking disparity from the predictions of the LAK theory. Measured values of the Kohler slope S range between 3×10^{-4} for a single-crystal sample to 3×10^{-2} for a pressed polycrystalline plate.

Several qualitative studies demonstrated that stress is a relevant parameter in the linear magnetoresistance of potassium. Single crystals, prepared and held under minimum-stress conditions, showed an average S value only 40% as large as for polycrystalline samples frozen in mineral oil. S was observed to increase monotonically with plastic strain, and this increase was reversed by annealing. The Kohler slope was found to be anisotropic: The [110] and [123] directions were found to have the smallest average values of S.

Due to the high reactivity of potassium, it was not possible to directly measure the effect of dislocations on S. It is impossible to determine from this experiment whether the linear magnetoresistance is caused by microscopic defects or by a strain-dependent band structure. Additional theoretical work will be required to provide a detailed mechanism for the linear magnetoresistance and its strain dependence.

[50] L. M. Falicov and P. R. Sievert, Phys. Rev. 138, A88 (1965).

ACKNOWLEDGMENTS

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APPENDIX

It is well known that a helicon surface mode exists. It produces a loss in the sample which is unrelated to bulk resistance.[51,52] It is important, of course, to ensure that such a loss does not affect the interpretation of the data reported above. It is shown in Ref. 52 that the effective Q measured will be related to the bulk loss ρ_{bulk} and a surface-mode parameter A by the relation

$$1/2Q=\rho/RB+A\,. \tag{A1}$$

It is important to note that A is a function only of the sample aspect ratio: the sample thickness divided by the sample diameter. The larger the value of this ratio, the larger A; the effect predominates when large areas of the samples are parallel to the field. Such a loss could be interpreted as linear magnetoresistance. The measured Kohler slope S_M is the sum of the bulk slope S_β and the surface effect A:

$$S_M=S_\beta+A\,. \tag{A2}$$

The experimental evidence that S_M depends on stress enables one to put an upper limit on A. All the samples used in this experiment were approximately the same size and so A would not be expected to vary significantly from sample to sample. S_M was found to vary by at least an order of magnitude with strain as a parameter. Since A is not expected to depend on stress, it is reasonable to conclude that $A\leq S_M$ minimum. The minimum S_M observed were in the region 3×10^{-4}, i.e., within the experimental error of the method. Thus, the value of A must be within the error of the method. It can be concluded that the method was not sufficiently sensitive to detect surface loss in the presence of bulk magnetoresistance. Thus, the conclusions about the validity of a bulk linear magnetoresistance stand.

[51] C. R. Legendy, Phys. Rev. 135, A1713 (1964).
[52] W. Schilz, Solid State Commun. 5, 503 (1967).