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Study of the Transverse Magnetoresistance of Polycrystalline Potassium†

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The impurity, strain, temperature, and size dependence of the transverse magnetoresistance of polycrystalline potassium has been investigated in fields up to 100 kG. The results of both low-field helicon experiments on large rectangular plate samples and four-terminal measurements on long wires show the existence of a nonlinear low-field behavior or "knee" previously reported in the magnetoresistance. The shape of the "knee" is observed to be strongly dependent on sample annealing and purity and, to a lesser extent, on temperature. In all samples under all experimental conditions, there is a linear magnetoresistance at high fields. The well-annealed wires exhibit a regular pattern in the impurity dependence of the high-field Kohler slope which differs from that reported for other simple metals. The Kohler slope is nearly independent of temperature below 4.2 K for samples of a wide range in purity. The experimental results are compared with predictions of macroscopic theories of a linear magnetoresistance due to sample geometry and inhomogeneities and with more recent proposals for an intrinsic linear magnetoresistance of potassium.

I. INTRODUCTION

Although theory predicts a saturating magnetoresistance for potassium, experimental work has produced a host of conflicting results. A linear magnetoresistance has generally been reported at high fields; however, there has been little agreement on the magnitude of the linear term and no satisfactory elucidation of the parameters determining its magnitude. Recently, there has also been disagreement as to whether there is a nonlinear low-field behavior or "knee" similar to that observed in the magnetoresistance of other closed-orbit metals such as indium and aluminum. While much of the uncertainty over early work centered on the effects of voltage probes and sample geometry, probeless methods and measurements in a wide variety of sample geometries have produced similarly conflicting results. Consequently, there has been increasing attention given to other factors, such as lattice defects and inhomogeneities, which could be of importance in a metal as soft and reactive as potassium.

Thus two primary reasons have led us to a paper of considerable length on measurements of the magnetoresistance of potassium. The first is the complicated nature of this phenomenon, which depends on several parameters and which has given rise to the variety of results having been reported. It requires a detailed analysis both to report our results and assess the extent to which they agree with previous work. The second reason is the crucial significance the magnetoresistance of potassium has for the semiclassical transport theory of metals. Potassium is generally regarded as one of the simplest metal systems, having a single band of conduction electrons with a nearly spherical Fermi surface. As such, it should have a saturating magnetoresistance at high fields. Yet there have been several intrinsic theories proposed in the last few years to explain the high-field linear magnetoresistance of potassium. We attempt to give a comprehensive assessment of these theories based on experimental data from a large number of samples. It is our hope that by providing this rather exhaustive "catalog" of experimental results we can give a sounder basis to future theoretical study of this important phenomenon.

In order to establish a basis for comparison with the work described in this paper, we briefly outline some of the more recent experimental results. Penz and Bowers[1] observed a linear magnetoresistance in the high-field regime $\omega_c\tau \gg 1$, where ω_c is the cyclotron frequency and τ the relaxation time determined from the zero-field resistivity.

Their probeless helicon measurements were done done at 4.2 K and above 10 kG on single-crystal samples having a residual resistivity ratio (RRR) between 1100 and 3900.[2] Characterizing the linear magnetoresistance by a slope in the Kohler variables $\Delta\rho/\rho_0$ and $\omega_c\tau$, they found no discernible pattern in the linear term with respect to its dependence on either sample purity or crystal orientation. The Kohler slopes ranged from 0.10×10^{-2} to 0.65×10^{-2} rad^{-1} over their 24 single-crystal samples. They did, however, find a qualitative dependence on the Kohler slope on strain; the slope always increased as the sample was stressed compressionally.

Subsequently, Babiskin and Siebenmann[3] have done four-terminal measurements on long (about 1 m) helically wound wires in a transverse magnetic field. These polycrystalline wires about 1 mm in diam were encapsulated in plastic tubes. In three such samples having RRR values of 560, 2230, and 4737 they observed a "knee" in the magnetoresistance extending as high as 15 kG in the purest wire. In each case a linear magnetoresistance was observed at high fields. Further, they claimed that the magnitude of the linear term decreased as the density and size of voids, which formed on the sample surface during solidification, were reduced. On this basis, they divided the magnetoresistance into an intrinsic saturating component and a linear component that was attributed to geometrical effects and nonuniform current distribution. They defined the saturating component $(\Delta\rho/\rho_0)_s$ to be the intercept found by extrapolating the linear portion of the magnetoresistance curve to zero field. We shall denote this intercept by $(\Delta\rho/\rho_0)_{H=0}$. Only for the lowest-purity sample do they present enough data to determine a Kohler slope; the value of 0.06×10^{-2} is about half of the lowest value reported by Penz and Bowers.

Jones[4] and Lass[5] have both done longitudinal and transverse four-terminal measurements on considerably shorter samples than those used by Babiskin and Siebenmann.[3] Neither report a knee at low fields. Although their experiments suffered from poor reproducibility, they found the Kohler slopes of both the longitudinal and transverse magnetoresistance to be generally larger but of the same order of magnitude as those of Penz and Bowers. Jones does not give the RRR of his samples but reports slopes in the range of $(2-5) \times 10^{-2}$ for both sodium and potassium. Lass observed smaller slopes from 0.3×10^{-2} to 1.1×10^{-2} for samples with RRR in the range 5000–6000.

Jones also investigated the dependence of the longitudinal magnetoresistance on extensional stress in wire samples. Unlike Penz and Bowers, he finds the Kohler slope to decrease (rather than increase) as the stress is applied; the magnitude of the effect is about the same.

In addition to four-terminal measurements, Lass measured the transverse magnetoresistance by a probeless induced torque method using both polycrystalline and unoriented single-crystal spheres of potassium (~ 1 in. diam). Kohler slopes inferred from these measurements were 1.4×10^{-2} and 2.6×10^{-2} for two single-crystal spheres (RRR $\approx$ 1200 and 2500, respectively); these values are more than twice as large as the largest slope reported by Penz and Bowers. Rotation studies showed no anisotropy in the magnetoresistance.

The original motivation for our experiment was to confirm the existence of a knee in the low-field magnetoresistance using the helicon method. In this way, the four-terminal measurements of Babiskin and Siebenmann[3] could be checked by a probeless technique in a different sample geometry and in samples subject to less strain. After some effort, we succeeded in observing a reproducible nonlinear magnetoresistance at low fields which was in qualitative, but not quantitative, agreement with Babiskin and Siebenmann. From this work, it seemed that the experimental understanding of the knee was at much the same level as that of the high-field linear term: There was qualitative agreement between experiments but an incomplete understanding of the dependence of the magnetoresistance on such sample conditions as impurity content, lattice defects, temperature, and size. We therefore began a systematic study of the magnetoresistance under these different sample conditions.

Since experiments on single crystals had shown the linear term to be more dependent on factors other than crystal orientation and since the low-field knee had been observed in polycrystalline samples, we chose to conduct this study using long polycrystalline extruded wires. In view of the past history of conflicting results, it was felt that a large number of samples should be measured. The relative ease in sample preparation, handling, and data analysis of the wire specimens permitted us to measure a large number of specimens under a wide variety of experimental conditions. Also, by using long wires we could obtain large voltage drops across the samples as well as minimize probe effects.

Experimental procedures for both the helicon and four-terminal measurements are presented in Sec. II. Section III contains the experimental results. The magnetoresistance of the helicon and unannealed wire samples is presented in Sec. IIIA. The effect of different sample treatments on the magnetoresistance and a comparison

TABLE I. Summary of data on the transverse magnetoresistance of the polycrystalline wire samples at 4.2 K[a].

Sample	Source	Diameter (mm)	Annealing time	RRR [b]	$10^2 S$ [c]	$(\Delta\rho/\rho_0)_{H_0}$ [d]	$(\Delta\rho/\rho_0)_{H=0}$ [c]
KX20	Alloy	1.5	24 h	136	0.47	0.006	0.001
KX21	Alloy	1.5	½ h	390	0.11	0.008	0.005
KX22	Alloy	1.5	½ h	690	0.16	0.019	0.013
			6 days	610	0.15	0.008	0.004
KX6	AC[e]	1.5	4 h	720	0.28	0.05	0.045
			~2 weeks	1110	1.36	0.14	−0.044
KX17	Alloy	1.5	23 h	790	0.76	Ref. f	−0.005
			6 days	670	0.43	Ref. f	0
KX7	AC	1.5	28 h	910	0.56	0.07	0.016
			22 days	1230	0.93	0.11	−0.067
KX10	AC	1.5	½ h	1970	0.49	0.17	0.118
			10 days	1390	1.01	0.20	−0.002
KX11	UMC[g]	1.5	28½ h	1100	0.76	0.15	0.092
			10 days	1660	1.30	0.26	0.040
KX8	MSA (I)[h]	1.5	26 h	1170	1.04	0.65	0.32
			11 days	2520	0.82	Refs. f, i	−0.05
KX24	Alloy	1.5	2 h	1350	0.14	0.02	0.006
			2½ days	3080	0.05	Refs. f, i	−0.013
KX15	MSA (I)	2.2	26½ h	1730	0.61	0.50	0.30
			14 days	2940	0.36	0.25	0.078
KX13	MSA (II)[h]	2.2	25 h	2440	0.31	0.18	0.087
			6 days	2960	0.39	0.31	0.018
KX18	MSA (II)	1.5	1 h	2540	0.19	0.11	0.066
KX12	MSA (II)	1.5	27½ h	3050	0.17	0.04[i]	0.018
			10 days	2760	0.30	Refs. f, i	−0.009
KX19	MSA (II)	1.2	28 h	3130	0.44	0.06[i]	0
			6 days	2530	0.61	Refs. f, i	−0.036
KX25	MSA (III)[h]	2.2	12 days	3240	0.42	0.41	0.075

[a]Samples are arranged in order of increasing RRR prior to annealing.

[b]$RRR = \rho(293\ K)/\rho(4.2\ K)$.

[c]The Kohler slope S is defined by $(\Delta\rho/\rho_0) = S\omega_c\tau + (\Delta\rho/\rho_0)_{H=0}$, where $\omega_c\tau = R_H H/\rho_0 = [R_H H/\rho(293\ K)]$ (RRR) $= 6.19\times10^{-4}$ (RRR)H. Here H is expressed in kG and we have used $R_H = -4.45\times10^{-9}$ Ω cm/kG (free-electron value); $\rho(293\ K) = 7.19\times10^{-6}$ Ω cm (from Ref. 9).

[d]Defined in the text.

[e]Allied Chemical Corp., Morristown, N. J.; Baker and Adamson Code 2080 ($\gtrsim$ 99% K).

[f]No "knee" observed.

[g]United Mineral and Chemical Corp., New York, N. Y. (99.99% K).

[h]MSA Research Corp., Evans City, Pa. (99.97% (K); (I) batch purchased June, 1966; (II) batch purchased February, 1970; (III) batch purchased May, 1970.

[i]Sample had a negative magnetoresistance at low fields.

with previous experiments is given in Sec. III B. By considering only the annealed samples, regularities in the impurity dependence of the magnetoresistance were observed; these are discussed in Sec. III C. The influence of impurity content and annealing on the temperature dependence is described in Sec. III D. The effect of sample size on the low-field magnetoresistance is summarized in Sec. III E. Finally, Sec. IV assesses the extent to which existing theories can explain our experimental observations. Our results are compared with the predictions of existing macroscopic theories for the linear term based on sample geometry and inhomogeneities, as well as with the intrinsic theories which have been proposed recently.

II. EXPERIMENTAL TECHNIQUES

A. Four-Terminal Measurements

Polycrystalline wire samples about 1 m long and from 1 to 2 mm in diam were used in this study. The wires ranged in purity from a RRR of 130–3130.[2] The lowest-purity samples were obtained by alloying Mine Safety Appliances Co. (MSA) potassium having a RRR of approximately 3000 with small amounts of sodium (RRR ≈ 2700). For example, a 1% by weight solution of sodium in potassium gave a base alloy having a RRR ≈ 4. Intermediate purities (RRR between 1100 and 1600) were not obtained by this alloying procedure but by using potassium from different manufacturers. Further details are given in Table I and information on sample preparation and handling can be found elsewhere.[6]

The wires were extruded into dehydrated paraffin oil at room temperature and wound directly onto an eight turns per inch threaded micarta former. In this way, the potassium did not come into contact with air during the entire operation. The extruder was motor driven so as to achieve the uniform travel necessary to keep the long wires from breaking. The helix formed by the wire was about 18 mm in diam and 5 cm long. Current contacts were made at each end by wrapping the potassium wire around a brass screw and squeezing it between two brass washers. Although it was not possible to wind the soft potassium wire noninductively, pickup noise was reduced by winding one of the voltage leads around the former in series opposition to the wire sample. The voltage leads were attached to the potassium about 5 cm in from the current contacts (approximately the distance occupied by one turn). Actual contact was made by cleaning the end of a No. 30 copper wire, tinning it with mercury, and pressing it through the center of the potassium wire from underneath until it just showed on the outside. All leads were held in place on the former with epoxy. This contact configuration was adhered to rigidly for all of the samples in an effort to keep constant whatever unknown probe effects might exist. The potassium wire remained permanently mounted on the former; it was either placed under dry oil to anneal or stored in liquid nitrogen. Just prior to running, the former was fastened to the bottom of the sample rod where plug-in pins made electrical contact with wires leading to the top of the cryostat.

The electronics consisted of a constant current source (drift $<$ 0.05%/h) and a Keithley 148 nanovoltmeter (accuracy of about 1%). The nanovoltmeter output was displayed on a six-place integrating digital voltmeter. Data were taken pointwise in order to eliminate induced voltages due to field ramping. At each value of the magnetic field, data were taken for positive and negative sample current and field directions in order to average out any thermal and Hall voltages. In practice, the Hall component ranged from 1 to 3% of the total voltage, although in a few samples it was as large as 10%. Generally, we tried to work with 100 μV signals in order to reduce scatter in the data due to short-term thermal-voltage fluctuations and magnet noise. This permitted us to resolve deviations from linearity on the order of a few tenths of a percent. A typical sample current of 10 A produces a field of 20 G at the surface of a 2-mm wire so that self-field effects were negligible.

Most of our runs were done in a 1-in. bore superconducting solenoid having a maximum field of 55 kG. A few representative samples were also run in a $1\frac{1}{2}$-in. bore 100-kG superconducting magnet. Field homogeneity over the 2-in.-long helical sample was about 2% for the 55-kG magnet and 1% for the 100-kG magnet.

One problem inherent in magnetoresistance measurements on helical samples is the mixture of transverse ($\rho_\perp$) and longitudinal ($\rho_\parallel$) components of the resistivity due to the finite pitch of the winding. The longitudinal component enters as $\cos^2\varphi$ where φ is the angle (approximately 87° in our samples) between the magnetic field and the sample current. Assuming $\rho_\parallel/\rho_\perp < 2$, this correction is $<$ 0.5% and shall be ignored in the following discussion.

B. Helicon Measurements

The helicon samples made of MSA potassium were rectangular plates about $25\times20\times3$ mm. These large samples were formed from molten potassium by solidification between glass plates, rather than by cutting from a single-crystal boule. This was done in an effort to minimize strain introduced by handling. After some practice, we obtained samples with surfaces having none or few visible grain boundaries and which required no cutting. These were then annealed for 1 or 2 days in xylene at room temperature, etched to remove oxidation, and stored in liquid nitrogen. For measurement, they were loaded at liquid-nitrogen temperature into a close-fitting rectangular-shaped pick-up coil. The samples were held in place with paper wedged gently between the coil and sample surfaces in order to prevent movement and vibration.

The helicon measurements were done using a standard crossed-coil technique and essentially the same detection scheme as that used by Penz and Bowers.[1] Our measurements were complicated by the low resonant frequencies (~8 Hz at 1 kG) inherent when using thick samples and low magnetic fields. For example, care had to be taken to ensure that the integrator used to obtain the absorptive signal had a sufficiently long time constant.

When the condition $\omega_c\tau \gg 1$ is no longer satisfied, the resistivity is not proportional to the frequency-amplitude ratio at resonance but involves a correction for the finite width of the resonance (for $\omega_c\tau \ll 1$ the Q approaches a limiting value of 0.5).[7] This is a 20% effect at 1 kG in a sample of RRR = 3000. Since the Q could not be estimated accurately from the full width at half-maximum of the absorption resonance, we were limited to working with the purest samples in order that $\omega_c\tau$ be as large as possible. Further discussion of experimental details and analysis procedures has been given elsewhere.[7]

For several other reasons it was not felt profitable to do an extensive helicon study of the annealing and temperature dependence of the magnetoresistance. The upper limit of 14 kG in the 2-in. bore superconducting magnet that was used did not permit us to determine the high-field Kohler slope with great confidence. We also encountered the usual problem in a helicon experiment of ac-

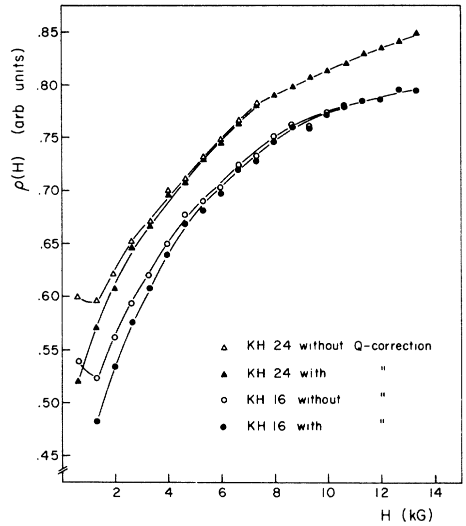

Fig. 1. Magnetoresistance curves for two helicon samples KH16 (RRR≈2500) and KH24 (RRR≈3100). Note that this is not a Kohler plot. The Q correction becomes negligible at the higher fields.

curately determining the zero-field resistivity. We estimated the RRR from the sample thickness and the Q of the resonance at 1 kG.[7]

III. EXPERIMENTAL RESULTS AND COMPARISON WITH PREVIOUS MEASUREMENTS

A. Evidence for a "Knee" at Low Fields: Helicon and dc Measurements

Figures 1 and 2 show some typical magnetoresistance curves for helicon measurements on annealed plates and dc measurements on unannealed wires. Note that neither figure is a Kohler plot. All of these samples have a nonlinear low-field magnetoresistance which is qualitatively similar to that reported by Babiskin and Siebenmann.[3]

The helicon measurements appear to have a larger magnetoresistance (higher knee) than either our wire samples or those of Babiskin and Siebenmann. However, for the helicon measurements, it is difficult to make a quantitative estimate of the magnetoresistance because of uncertainties in the zero-field resistivity and the size of the Q correction. The observed knee cannot be attributed to either nonlinearity or phase shifts in the detection scheme since even at 1 kG the helicon resonant frequency of 8–10 Hz is well within the bandpass of the amplifiers. Thus, we are confident of the existence of a nonlinear component in the magnetoresistance deduced from helicon measurements although we are reluctant to draw conclusions relating to its magnitude. The remaining discussion in this paper will, therefore, deal exclusively with measurements on long-wire specimens.

There are some features of the magnetoresistance of the unannealed wire samples (Fig. 2) which differ from results reported by Babiskin and Siebenmann.[3] The major difference is the magnitude of the magnetoresistance that we observe at the knee. Babiskin and Siebenmann report zero-field intercepts of the linear term $(\Delta\rho/\rho_0)_{H=0}$ ranging from 0.6×10^{-2} to 2.2×10^{-2} for the three wire samples mentioned previously. As can be seen in Fig. 2, many of the samples that we studied showed larger intercepts. Indeed, there were two unannealed samples KX8 and KX15

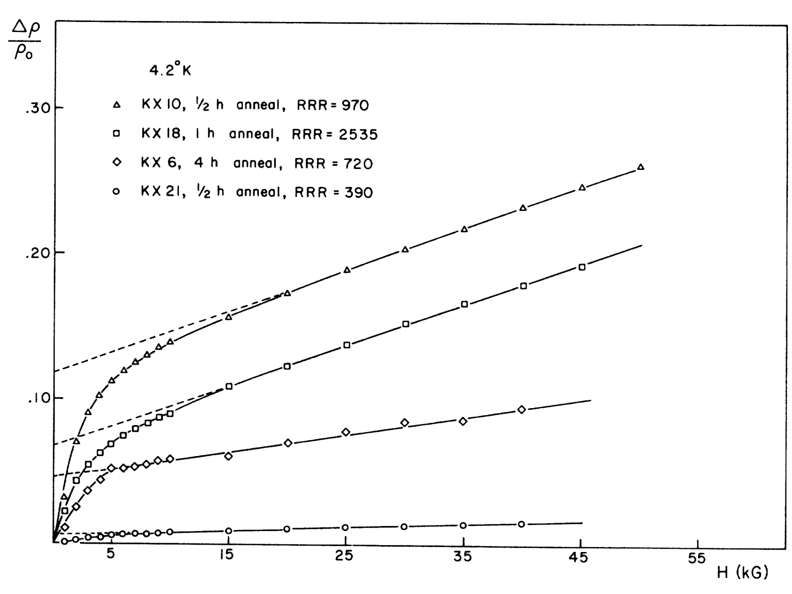

Fig. 2. Magnetoresistance curves $(\Delta\rho/\rho_0)$ vs H for some typical unannealed ($\lesssim$ 4 h) wire samples.

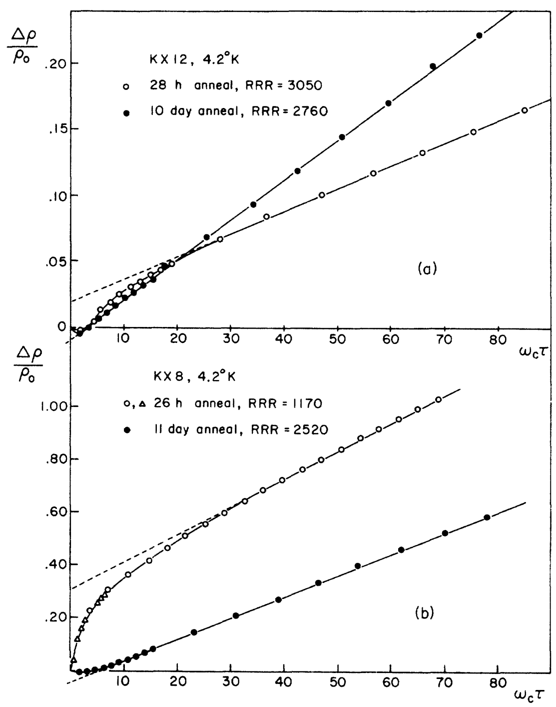

Fig. 3. Effect of annealing on the magnetoresistance of some high-purity wire samples: (a) no deviation from linearity at low fields and the Kohler slope increases; (b) again no deviation from linearity at low fields but the Kohler slope decreases. The points marked Δ were taken in the 55-kG magnet; those marked o were taken in the 100-kG magnet 10 days later after storing the sample in liquid nitrogen.

with $(\Delta\rho/\rho_0)_{H=0}$ as large as 30×10^{-2} [see Figs. 3(b) and 4(b)].

Another difference that we observe is related to the impurity dependence of the magnetoresistance. Babiskin and Siebenmann[3] found that $(\Delta\rho/\rho_0)_{H=0}$ increased and the knee region encompassed a broader field range as the sample purity (RRR) increased. While we observe this pattern to hold for the three lowest-purity samples in Fig. 2, this same figure shows that both $(\Delta\rho/\rho_0)_{H=0}$ and the width of the knee decreased when the RRR increased from 970 (KX10) to 2535 (KX18).

B. Effect of Sample Treatment on Magnetoresistance

All the wire samples classed as unannealed were run within 1 day of extrusion. The strain dependence of the magnetoresistance[1,4] reported previously suggested to us that the differences between our low-field results and those of Babiskin and Siebenmann[3] might be due to defects introduced during extrusion. To check this possibility, measurements were made on 12 of the 16 wire samples after they had been annealed. By annealing, we mean that the wire, mounted on the former, remained immersed in dehydrated paraffin oil, under vacuum, and at room temperature for a period ranging from 3 days to 3 weeks. It was difficult to achieve longer annealing times since this could result in a decreased RRR of the specimen, oxidation of the current and voltage contacts, and eventual loss of electrical continuity.

Table I summarizes the effect of annealing on those properties of the magnetoresistance which can be described quantitatively in some consistent manner. However, the variety of behavior associated with annealing is best demonstrated by the magnetoresistance curves. The samples fall roughly into three groups according to the RRR prior to annealing: the purest samples in Figs. 3 and 4, the intermediate-purity samples in Figs. 5 and 6, and the lowest-purity samples in Fig. 7. Within these groups, the samples could be further divided according to whether the Kohler slope increased or decreased upon annealing.

A common effect of annealing pure samples is illustrated in Fig. 3[8]; the knee observed in the unannealed state of KX8 and KX12 disappeared entirely after annealing. Samples KX19 and KX24, which behaved similarly, are shown in Figs. 8 and 14, respectively. In these four annealed samples, a negative magnetoresistance immedi-

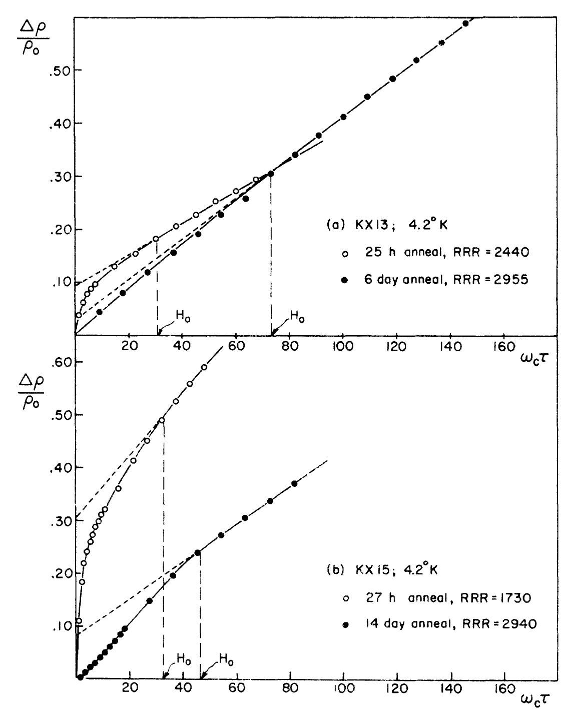

Fig. 4. Effect of annealing on some other high-purity wires: There is still a small deviation from linearity at low fields and the Kohler slope (a) increases and (b) decreases.

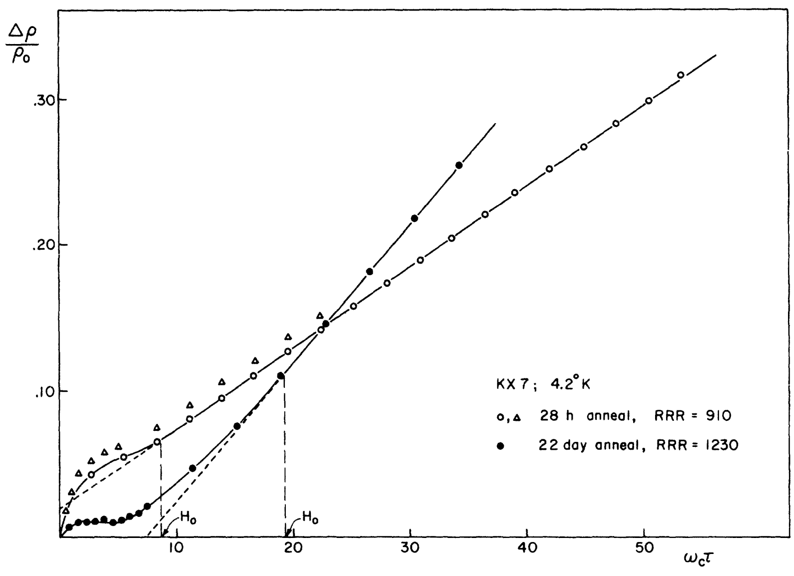

Fig. 5. Effect of annealing on the magnetoresistance of an intermediate-purity sample: Quasisaturation develops at low fields and the Kohler slope increases. Measurements were made on the unannealed sample in the 55-kG magnet (Δ) and then in the 100-kG magnet (o) after storing for 36 days in liquid nitrogen.

ately precedes the linear term; this results in a negative value for $(\Delta\rho/\rho_0)_{H=0}$. The effect of annealing on the Kohler slope of these samples ranged from decreasing by almost a factor of 3 for KX24 to nearly doubling for KX12. Note that annealing increased the RRR by almost a factor of 2 for the two samples (KX8 and KX24) in which the Kohler slope decreased. Figure 3(b) also illustrates the high degree of reproducibility that we were able to achieve with the wire samples. Before annealing, KX8 was run in the 55-kG magnet, stored in light nitrogen for 10 days, and then run again in the 100-kG magnet. As is shown by the unannealed curve in Fig. 3(b), there was no detectable change in the magnetoresistance in these two different runs.

Some vestige of a knee remained after annealing two other high-purity samples of larger diameter. Neither of these samples showed a negative magnetoresistance at low fields. As shown in Fig. 4(a), the Kohler slope of sample KX13 increased with annealing. In addition, a very small deviation from linearity was observed below 50 kG ($\omega_c\tau = 70$). Figure 4(b) shows a similar but more pronounced effect in sample KX15. Here a linear magnetoresistance is observed at low fields, an abrupt change in Kohler slope takes place around 24 kG, and a second linear region is present at high fields. Again, we note that for KX15 the decrease in the Kohler slope was accompanied by

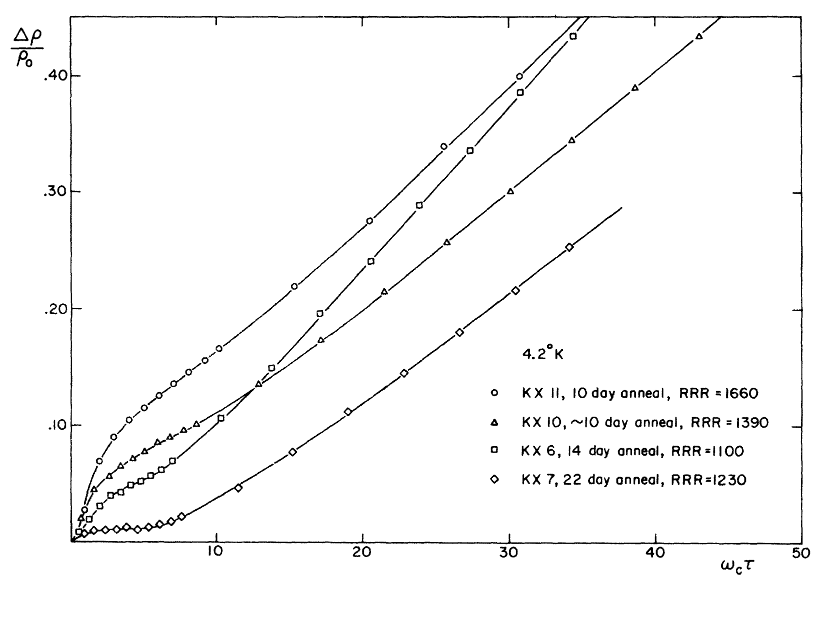

Fig. 6. Magnetoresistance of some intermediate-purity wire samples for increasing annealing times.

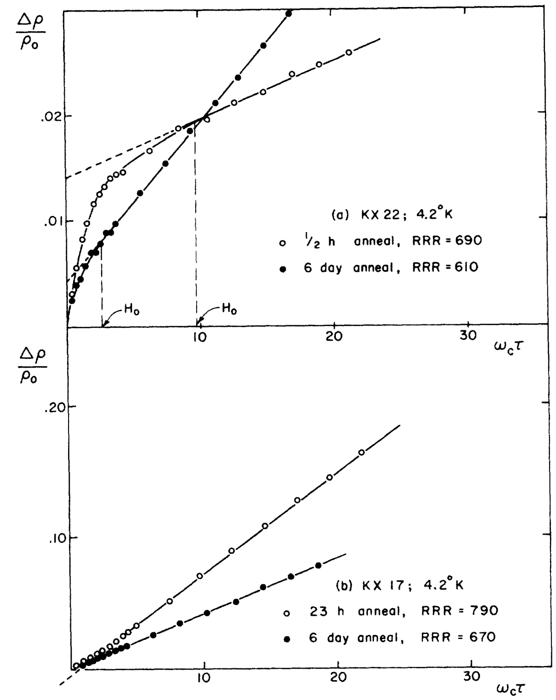

Fig. 7. Effect of annealing on some Na-K alloy samples showing the Kohler slope (a) to increase and (b) to decrease.

a large increase (factor 2) in the RRR.

Samples of intermediate purity, that is a RRR ranging from 1100 to 1700 prior to anneal, showed yet a different change in low-field behavior upon annealing. Figure 5 shows the most extreme case, that of KX7; the magnetoresistance in the annealed state increases about 1% in the first few kilogauss and then levels off in "quasisaturation." At 6 kG, the magnetoresistance again increases until the linear term dominates above about 20 kG ($\omega_c\tau = 20$). Comparing samples KX6, KX7, KX10, and KX11 shown in Fig. 6, the degree of quasisaturation appears to be related to the annealing time. The largest increase in Kohler slope of any of the wire samples, more than a factor of 4, was observed in sample KX6. For other samples of intermediate purity, the Kohler slope about doubled and the RRR (including KX6) increased by about 50%. [All of these samples were made from Allied Chemical Corp. (AC) potassium except for KX11, which was of United Mineral and Chemical Corp. (UMC) stock.]

The lowest-purity samples showed the least qualitative change upon annealing. The magnetoresistance curve of KX22 shown in Fig. 7(a) retained basically the same shape although annealing reduced both the height and width of the knee and increased the Kohler slope. Sample KX17 shown in Fig. 7(b) had a small negative zero-field intercept before the anneal (but no negative magnetoresistance). After annealing, there was no detectable deviation from linearity even at low fields.

Such a wide variety of behavior makes it very difficult to summarize the effect of annealing. We can say that annealing always resulted in a decrease in the low-field magnetoresistance. This is manifested in several ways including the complete disappearance of the knee, the observation of quasisaturation, and the extension of a linear term to very low fields. These profound effects produced by annealing, particularly the negative values of $(\Delta\rho/\rho_0)_{H=0}$, make a consistent definition of an intrinsic "saturating component" as introduced by Babiskin and Siebenmann difficult to apply. Further discussion of this point is contained in Sec. IV A.

In an effort to obtain a better description of the magnetoresistance of our samples, we found that some significance could be attached to the field at which the linear term enters. The magnetoresistance at this onset field H_0 is denoted by $(\Delta\rho/o_0)_{H_0}$. The onset field can be determined from the magnetoresistance curves of those samples that exhibit a knee: H_0 is that field where the

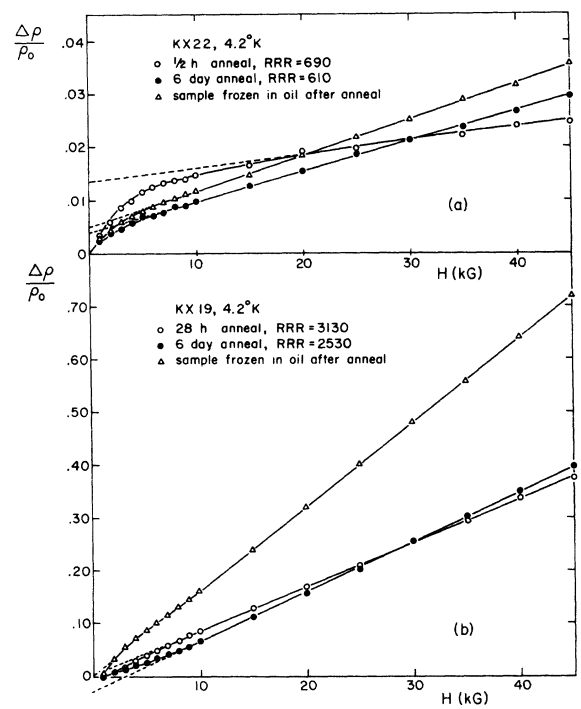

Fig. 8. Effect of freezing oil onto a wire sample; comparison with magnetoresistance before and after annealing. (a) Alloy wire sample of Fig. 7(a); (b) high-purity wire sample. Note that these are not Kohler plots.

low-field extrapolation of the linear magnetoresistance differs from the actual magnetoresistance by more than the experimental error. This prescription has some inherent difficulties in that frequently the approach to the high-field linear behavior is very slow. Also, there is the error in interpolating between data points. The definition of the onset field is illustrated for different characteristic shapes of magnetoresistance curves in Figs. 4, 5, and 7(a).

The decrease in the low-field magnetoresistance can now be demonstrated by comparing $(\Delta\rho/\rho_0)_{H_0}$ before annealing with the magnetoresistance at the same value of $\omega_c\tau$ after annealing [see Figs. 4, 5, and 7(a)]. Note that because of the usual large increase in H_0 with annealing, there is no assurance that $(\Delta\rho/\rho_0)_{H_0}$ will decrease with annealing. This is clear from the data in Table I. The usefulness of these parameters in characterizing the low-field magnetoresistance will become more clear in discussions of the impurity and temperature dependence of the magnetoresistance in the following sections.

As for the effect of annealing on the high-field linear term, the Kohler slope could either increase or decrease. Only for samples of intermediate purity shown in Fig. 6 did the Kohler slope (and the RRR) consistently increase with annealing. In Sec. IV we will show how the change in Kohler slope with annealing can be correlated with the accompanying change in the RRR. As a result of annealing, the range in Kohler slopes nearly doubled; for the unannealed samples Kohler slopes ranged from 0.06×10^{-2} to 1.0×10^{-2} (a factor of 17) while for the annealed samples they ranged from 0.05×10^{-2} to 1.4×10^{-2} (a factor of 28).

Lass[4] is the only one to report an annealing experiment similar to ours. His induced torque measurements on single-crystal spheres were repeated after the samples had annealed in oil for 140 days. In both of these samples, the RRR about doubled and the Kohler slope decreased by about 25%. Except for our larger decreases in the Kohler slope, this is very similar to the behavior that we observed in samples KX8, KX15, and KX24 (Kohler slopes decreased by 25, 80, and 300%, respectively).

The "matchstick"-sized sample (RRR $\approx$ 5500) that was used by Lass[4] for four-terminal measurements of the transverse magnetoresistance showed qualitatively the same behavior as our purest annealed samples KX8, KX12, KX19, and KX24 for which the linear term immediately follows an initial negative magnetoresistance. Lass[4] found that the Kohler slope varied by a factor of 3 (from 0.32×10^{-2} to 0.92×10^{-2}) depending where on the sample the voltage contacts were made. These values are in the range of slopes of our pure samples (except for KX24 which had the shortest annealing time).

It is not clear to what extent the samples of Babiskin and Siebenmann[3] may be considered annealed. By avoiding the extrusion process, they have obtained samples consisting of a few large crystallites. However, the defects introduced into the sample by encapsulation in a plastic tube are not known. They did not report any change in the magnetoresistance below the knee as the linear component was reduced by minimizing the number of voids on the surface of the sample. At first sight, the decrease in the low-field magnetoresistance of our samples with annealing might be regarded as consistent with the generally small zero-field intercepts of their linear term. Yet, at 4.2 K, they do not mention observing effects such as quasisaturation and the absence of a knee altogether.

In an attempt to relate our annealing studies to the conflicting strain dependence reported by Penz and Bowers[1] and Jones,[3] we made some qualitative strain-dependence measurements by covering samples with paraffin oil at room temperature and then quenching them in liquid nitrogen. The results of such measurements on two samples KX22 and KX19 are given in Fig. 8. Values for the RRR and Kohler slope are not tabulated since it obviously was not possible to perform a room-temperature measurement of the resistivity with the frozen oil in place (to determine the Kohler slope and RRR requires knowing ρ_0).

We find that a coating of frozen oil enhances the over-all magnetoresistance and increases the high-field slope of the linear term. This is in agreement with the observations of Penz and Bowers for oil-coated helicon specimens. In the low-purity sample KX22 the knee is lower than in the preannealed state. However, in the high-purity specimen KX19, $(\Delta\rho/\rho_0)_{H_0}$ exceeds the unannealed value. This effect raises the question of whether by encapsulating their wire samples, Babiskin and Siebenmann enhanced the low-field magnetoresistance of their specimens. This may be the reason why they observe a knee in pure samples, whereas in general we do not.

Two other interesting effects were observed which may explain the origin of the enhanced magnetoresistance when the sample is frozen in oil. First, we observe a *decrease* in the zero-field resistance of KX22 by 14% and in KX19 by 47%. These samples had a RRR of 610 and 2530, respectively, in their annealed state prior to coating with oil. Second, when KX22 was warmed to room temperature, the oil removed, and then *immediately* rerun, the magnetoresistance at 15 and 35 kG was equal to that observed previously in the *annealed* state. These observations suggest that freezing oil onto the sample did not introduce defects. Instead, the differential thermal contraction between the oil and potassium produced a nearly hydrostatic pressure on the sample.

A decrease in the zero-field resistivity with pressure is well known in potassium and has been studied most recently by Dugdale and Gugan.[9,10] If we use their value of $\partial \ln \rho_i'/\partial \ln V = 10.7 \pm 1$ at 4.2 K, where ρ_i' is the ideal resistivity of potassium at constant density, then we see that a 50% reduction in resistivity is not unreasonable for a 5% decrease in sample volume. This is the approximate reduction in volume between 293 and 77 K which we observe for paraffin oil alone. Also the smaller reduction in zero-field resistivity for the low-purity sample KX22 can be understood since this specimen requires a larger correction for the volume dependence of the residual resistivity.

In contrast to these observations, Jones reported that the zero-field resistivity of his wires *increased* under extensional stress. This suggests that, unlike the frozen-oil experiments, a significant number of defects were introduced into his samples.

Besides using frozen oil, Penz and Bowers tried straining their samples by applying compressional stress through a spring-loaded plunger. This treatment also increased the Kohler slope. Because the specimens were plastically deformed during this process, it is not clear whether the volume or just the thickness of the sample decreased. Whether a compressional stress increased or decreased the RRR is also not known, since ρ_0 was determined by extrapolation of the linear term in the magnetoresistance to zero field.

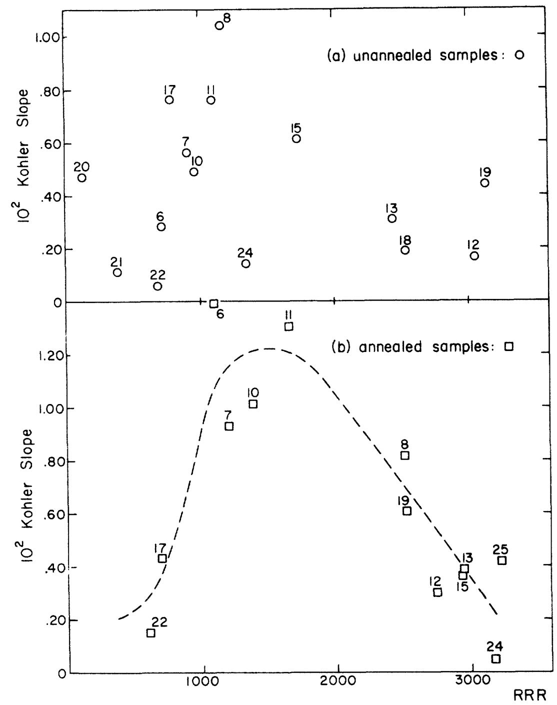

Fig. 9. Kohler slope as a function of sample RRR for (a) the unannealed and (b) the annealed wires.

C. Impurity Dependence of Magnetoresistance in Well-Annealed Samples

Section III B examined the effect of annealing upon the magnetoresistance of some representative samples. In this section, instead of following the history of one sample, the annealed samples will be considered as a group in order to study the impurity dependence of the magnetoresistance.

If Kohler's rule were obeyed, the Kohler slope would be independent of the RRR of the sample. However, our wire samples exhibit a large variation in Kohler slope with RRR at 4.2 K. In Fig. 9 we have plotted the Kohler slope as a function of RRR for (a) the unannealed and (b) the annealed samples. We find a significant reduction in the scatter of the points for well-annealed samples. The pattern which emerges reveals a maximum in the Kohler slope for samples having an intermediate purity. For low purities there is a rapid increase in the Kohler slope, and at high purities the Kohler slope decreases almost inversely with RRR. There is some indication that the Kohler slope approaches a value of about 0.4×10^{-2} for RRR > 2800 in the best annealed samples.

Since such a pattern in the impurity dependence of the Kohler slope of potassium has not been observed before, some internal check on this pattern would be very helpful. Close examination reveals that the pattern in Fig. 9(b) is fairly successful in explaining the change in Kohler slope that occurs upon annealing in terms of the accompanying change in RRR. The decrease in Kohler slope of KX8 and KX15 with annealing follows the trend given by the dashed curve for an increase in RRR from an intermediate to a high purity. The increase in Kohler slope of samples KX6, KX7, KX10, and KX11 also follows the dashed curve in Fig. 9(b) for an increase in RRR from a low to an intermediate value. In fact, KX13 and KX22 are the only two samples for which the sign of the change in Kohler slope cannot be accounted for by the observed change in RRR, and for these samples the RRR changed by a relatively small amount. Thus for our annealed samples at 4.2 K, the RRR appears to be the most important factor in determining the Kohler slope.

Deviations from Kohler's rule have also been observed in other simple metals such as In[11,12]

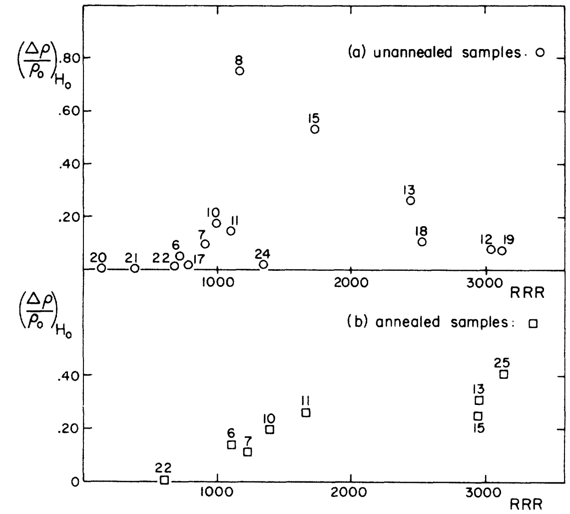

Fig. 10. Magnetoresistance $(\Delta\rho/\rho_0)_{H_0}$ at the onset field H_0 of the linear term as a function of sample RRR for (a) the unannealed and (b) the annealed wires. The high-purity annealed samples which showed no knee (Fig. 3) are not included in (b).

and Al,[13] which have a high-field linear term in the transverse magnetoresistance. No one has reported a maximum in the Kohler slope as a function of RRR at fixed temperature. Generally in these metals, an increase in RRR produces an over-all enhancement of the magnetoresistance and a slight increase in Kohler slope at 4.2 K.

While the pattern in Fig. 9(b) enables one to correlate changes in Kohler slope with changes in RRR resulting from annealing, our discussion up to now has begged the question of why these changes occur. In some samples, we did observe a growth in crystallite size during annealing. Although grain boundaries were never observed immediately after extrusion, annealing for a week produced crystallites as long as 1 cm in a few samples. The growth in crystallite size is probably an indication that internal defects in the specimen are decreasing in number during annealing.

However, in some samples, it is difficult to explain such large increases in RRR solely on the basis of a reduction in the number of dislocations. In KX24, the most dilute of the Na-K alloys, the RRR more than doubled after annealing only $2\frac{1}{2}$ days. It is possible in those samples for which a large increase in RRR is observed upon annealing that the (dominant) Na impurity atoms are precipitating or clustering in such a way as to reduce the residual resistivity.[14] Such a hypothesis is consistent with the large diffusion coefficient of sodium in potassium reported by Barr *et al.*[15]

Perhaps the most convincing evidence for impurity diffusion is that the RRR could decrease as well as increase after annealing in oil. For instance, KX19 was one of the samples which showed a visible increase in crystallite size but in which the RRR fell 20%, from 3130 to 2530. The RRR of samples KX12, KX17, and KX22 also decreased upon annealing.

If it is assumed that the pattern in the RRR dependence of the Kohler slope in the annealed samples resulted from a reduction in the density of lattice defects, then it should be possible to use Fig. 9(b) to separate the defect dependence from the impurity dependence of the Kohler slope. To pursue this idea, consider the Kohler slope for unannealed samples. Except for KX20, the lowest-purity alloy sample, the Kohler slope for the unannealed specimens is less than or roughly equal to those for annealed samples of comparable purity. This implies that the effect of reducing the number of defects by annealing is to increase the Kohler slope. The decrease in Kohler slope which Jones observed in strained specimens is consistent with such an interpretation.

It is much more difficult to characterize the

TABLE II. Temperature dependence of the magnetoresistance for the polycrystalline wire samples.

Sample	T (K)	RRR (T)[a]	Annealing time	$10^2 S$	H_0(kG)	$(\Delta\rho/\rho_0)_{H_0}$	$(\Delta\rho/\rho_0)_{H=0}$
KX6	4.2	1153	~2 weeks[b]	1.51	17	0.14	−0.044
	3.3	1202	2 weeks	1.51	17	0.14	0.044
	2.0	1228	2 weeks	1.51	17	0.14	0.044
KX13	4.2	2955	6 days	0.40	49	0.31	0.018
	1.6	3360	6 days	0.40	48	0.34	−0.010
KX25	4.2	3236	12 days[c]	0.42	44	0.40	0.075
	3.3	3590	12 days	0.44	21	0.23	0.028
	2.5	3706	12 days	0.44	16	0.18	0.020

[a] RRR $(T)=\rho(293\text{ K})/\rho(T\text{ K})$. .
[b] Sample had been stored for 3 months in liquid nitrogen after having been run in the annealed state (see Table I).
[c] Sample handled roughly after annealing.

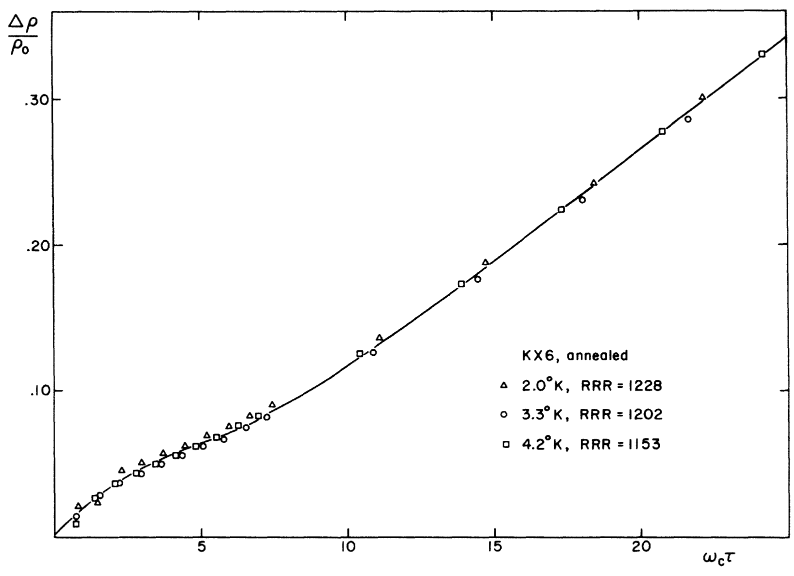

Fig. 11. Temperature dependence of an annealed intermediate-purity wire sample. The magnetoresistance obeys Kohler's rule as the temperature is reduced.

low-field magnetoresistance of annealed samples by a single parameter such as can be done at high fields by using the Kohler slope. The magnetoresistance at the onset field of the linear term, $(\Delta\rho/\rho_0)_{H_0}$ is shown as a function of RRR in Fig. 10. Those samples which showed no deviations from linearity have, of course, not been included. In annealed samples, it is evident that $(\Delta\rho/\rho_0)_{H_0}$ tends to increase monotonically with RRR. This trend in the low-field behavior is not inconsistent with the increase in $(\Delta\rho/\rho_0)_{H=0}$ with RRR observed by Babiskin and Siebenmann.

The large value of $(\Delta\rho/\rho_0)_{H=0}$ in our purest specimens is due to the wide field range of a nearly linear magnetoresistance below H_0 (see Fig. 4). This is a behavior distinctly different from quasisaturation which is observed in specimens having a somewhat lower RRR. Annealing served to accentuate these two characteristic forms of low-field behavior. The lowest-purity sample KX22 for which the over-all magnetoresistance is greatly suppressed had qualitatively the same magnetoresistance curve after annealing and the smallest value of $(\Delta\rho/\rho_0)_{H_0}$.

The monotonic impurity dependence of $(\Delta\rho/\rho_0)_{H_0}$ characterizing the low-field magnetoresistance contrasts with the maximum observed in the high-field Kohler slope as a function of RRR [Fig. 9(b)]. Another distinction between the high- and low-field magnetoresistance can be drawn on the basis

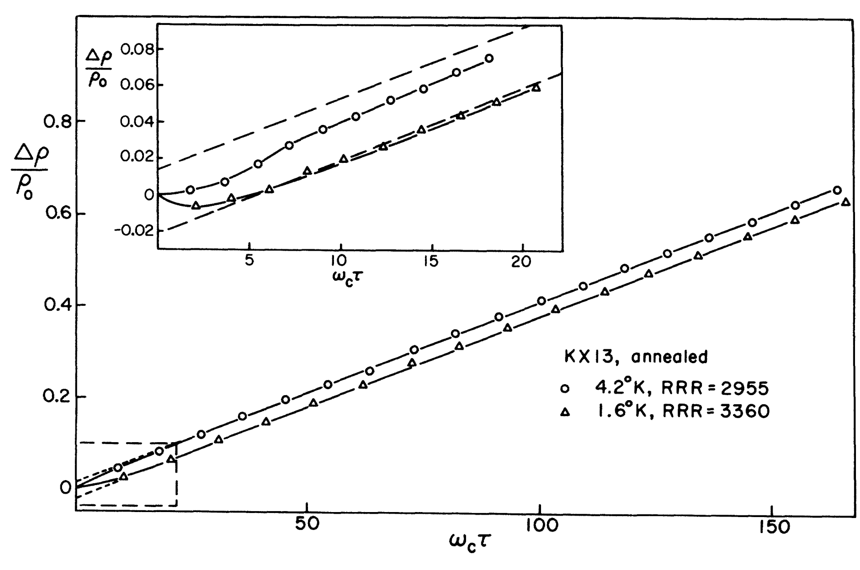

FIG. 12. Temperature dependence of the annealed high-purity wire sample in Fig. 4(a). The insert contains data from low-field measurements in a different magnet. The dotted lines are an extrapolation of the high-field linear magnetoresistance.

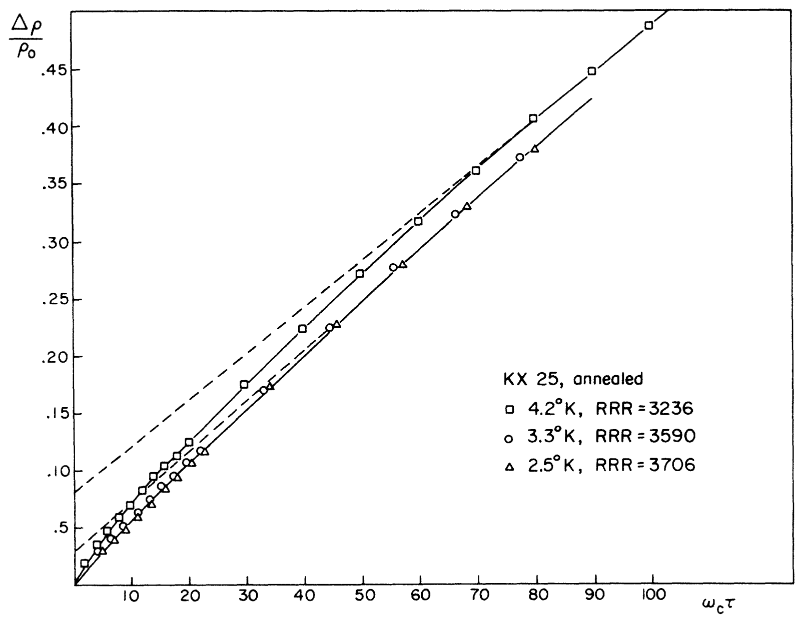

Fig. 13. Temperature dependence of a high-purity sample which had been handled roughly after annealing. The lower curve is the best fit to the data at 3.3 and 2.5 K.

of the effect of annealing. The low-field magnetoresistance always decreased with annealing, regardless of the change in RRR, while at high fields we have noted the correlation between changes in RRR and Kohler slope. This may indicate that much of the low-field magnetoresistance, particularly in intermediate- and high-purity samples, is caused by lattice defects. However, in Sec. III D evidence is presented which shows that defects cannot be the only cause of the nonlinear behavior in pure specimens.

D. Temperature Dependence of Magnetoresistance

The temperature dependence of various parameters which characterize the magnetoresistance is summarized for three samples in Table II. KX6 was selected for temperature measurements since in the annealed state it had the largest Kohler slope but only an intermediate RRR. It was thought that if an electron-phonon scattering mechanism were responsible for the linear magnetoresistance, then we might find a large decrease in the Kohler slope with temperature but only a small decrease in the zero-field resistivity. However, as shown in Fig. 11, KX6 obeys Kohler's rule almost perfectly as the temperature was reduced. Despite the fact that the zero-field resistivity changed by 7% between 4.2 and 2 K, the Kohler plots for these two temperatures coincide to within 1%, the experimental accuracy.

A purer sample, KX13 (RRR = 2955), exhibited a different behavior. As shown in Fig. 12, at 1.6 K a negative magnetoresistance developed at the lowest fields while at higher fields the magnetoresistance dropped uniformly by about 3%. Despite the 14% reduction in the zero-field resistivity between 4.2 and 1.6 K, the Kohler slopes remained equal at the two temperatures. A more detailed analysis in which we subtracted out the high-field linear term from the magnetoresistance showed that the onset fields of the linear term were about equal at the two temperatures, $H_0 \approx 50$ kG.

The greatest variation with temperature was observed in another pure sample, KX25 (RRR = 3236). In Fig. 13, we see that the onset field and $(\Delta\rho/\rho_0)_{H=0}$ decrease by a factor of 2 between 4.2 and 3.3 K. However, the Kohler slopes are again nearly equal for the temperatures investigated. The high knee characterized by the large value of $(\Delta\rho/\rho_0)_{H=0}$ at 4.2 K suggests that the sample may have been strained (recall that one expects the low-field magnetoresistance to decrease with annealing). It is possible that the relatively long annealing time was not sufficient to remove all defects. A similarly large zero-field intercept and strongly temperature-dependent H_0 were observed in KX13 when the magnetoresistance was remeasured after some rough handling that was necessary in order to restore electrical continuity. This behavior suggests that straining the sample enhances the temperature dependence of the magnetoresistance.

The only other recent temperature-dependence measurements of the magnetoresistance have been reported by Babiskin and Siebenmann.[3] In contrast to our wire sample KX6 (RRR = 1153), their lowest-purity sample (RRR = 560) showed a measurable temperature dependence: $(\Delta\rho/\rho_0)_{H=0}$ decreased from 0.6×10^{-2} at 4.2 K to 0.4×10^{-2} at 1.4 K with only a 3% decrease in the zero-field resistivity.

Their purest sample (RRR = 4437) showed a negative magnetoresistance at 1.4 K which completely masked any knee. This generally greater dependence of the magnetoresistance on temperature might be related to the more pronounced temperature dependence that we observe in our annealed samples which have been subsequently strained. In the measurements of Babiskin and Siebenmann, thermal contraction of the plastic encapsulating tube is a potential source of strain. In fact if voids, particularly surface voids, are present, then differential thermal contraction (the plastic is known to contract more than the potassium) will cause the potassium to flow plastically into the void regions. Babiskin and Siebenmann do not include a discussion of the temperature dependence of the high-field linear term.

E. Negative Magnetoresistance and Size Effect

A negative magnetoresistance was observed in the smallest diameter samples ($d \lesssim 1.5$ mm) of the purest potassium, RRR > 2500. The "free-electron" mean free path (MFP), $l = mv_F/ne^2\rho_0$, is about 0.08 mm for a specimen having a RRR = 2500. A negative magnetoresistance of less than 2% in these samples is consistent with a rough estimate of the size-effect contribution to the zero-field resistivity of order $l/d \approx 5\%$. In samples KX12 and KX19 a negative magnetoresistance occurred prior to annealing but was not particularly enhanced by annealing (the RRR decreased in both cases). KX8 and KX24 only showed a negative magnetoresistance after annealing. In both of these samples annealing was accompanied by a large increase in RRR. Thus, for the limited range of sample diameters considered here, a minimum RRR appears to be the most important factor for the existence of a negative magnetoresistance.

We did not obtain more quantitative evidence that the negative magnetoresistance correlated with wire diameter and zero-field resistivity. Among the samples in Fig. 14, the lowest-purity specimen KX8 had the largest negative magnetoresistance, while the sample of smallest diameter, KX19, showed the smallest negative magnetoresistance. These two observations are contrary to what one would expect for a conventional size effect.

In the case of KX24, the sample with the smallest Kohler slope, the negative magnetoresistance persists up to 15 kG before the linear term begins to dominate. It is somewhat surprising that the resistance minimum occurs at such a high field. Even if the linear term were as large as for KX8, the resistance minimum would still be at 2 kG or about twice as high as in the other samples. This disparity cannot be accounted for on the basis of sample size or electron MFP, since all of the samples in Fig. 14 are of comparable purity and, except

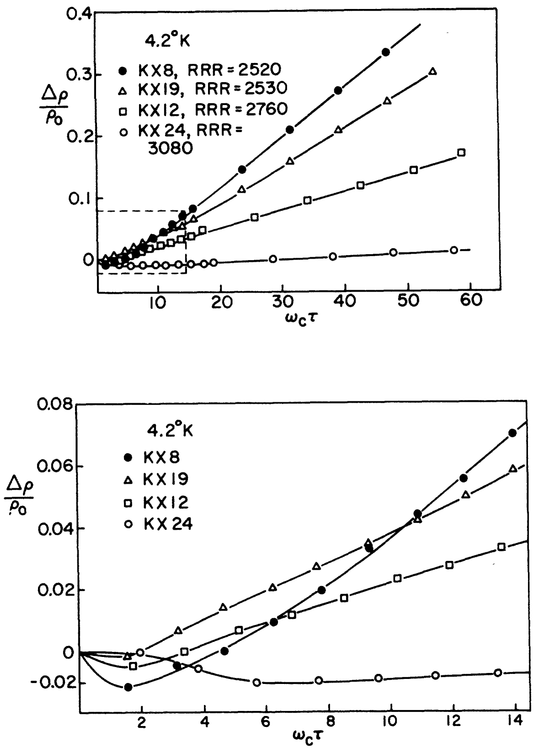

Fig. 14. High-purity annealed wires showing a negative magnetoresistance.

for KX19, all had the same diameter. In previous size-effect experiments in a transverse magnetic field on much thinner wires,[16,17] the field at the resistance minimum essentially scaled inversely with sample diameter.

In our wire samples, it is unlikely that the high-field linear term was affected by sample diameter. Although we have seen how the Kohler slope decreases at high purities, a similar dependence is not observed among the wire samples of comparable purity but decreasing diameter.

F. Summary of Experimental Results

The nonlinear low-field magnetoresistance which we have observed by both helicon and four-terminal dc techniques depends strongly on both sample annealing and purity and, to a lesser extent, on temperature. Depending on how long a sample has been annealed, we can obtain, without altering the electrical connections to the specimen, qualitatively different magnetoresistance curves. For example, in the high-purity samples the "knee" can vanish completely upon annealing. In annealed samples of intermediate purity we observe a new effect, that of quasisaturation.

By restricting our attention to annealed samples, we have recognized patterns in the impurity dependence of the magnetoresistance. The Kohler slope has a maximum as a function of the specimen RRR. In those samples which have a knee, the magnetoresistance at the onset of the linear term, $(\Delta\rho/\rho_0)_{H_0}$, increases monotonically with RRR. The existence of these patterns indicates that the magnetoresistance of well-annealed samples is more reproducible. There is some evidence that a well-annealed sample is characterized by a growth in crystallite size and a decrease in the density of lattice defects.

We have attempted to use these patterns to separate the impurity from the strain dependence of the high-field magnetoresistance. The annealing studies indicate that the Kohler slope depends most strongly on the RRR of the specimen. If strain or defects do have an effect on the linear term, it is to decrease the Kohler slope. This conclusion is supported by the strain dependence of the linear term in the longitudinal magnetoresistance observed by Jones. We have proposed that the conflicting strain dependence reported by Penz and Bowers in the transverse magnetoresistance could be explained by a decrease in sample volume under the application of the "stress" and is therefore really a pressure effect.

Defects appear to have just the opposite effect on the low-field magnetoresistance. Below the knee, the magnetoresistance always decreased with annealing giving rise to a variety of low-field behavior. In the annealed samples the height and width of the knee increased with sample RRR.

The low-field magnetoresistance is more strongly temperature dependent than the high-field linear term. The low-field magnetoresistance always decreases with temperature while the high-field Kohler slope is nearly independent of temperature. There exists some evidence that introducing many defects into the samples enhances the temperature dependence.

We associate the presence of a negative magnetoresistance in our pure small-diameter wires with a size effect. However, one of the samples, KX24, had a negative magnetoresistance which was sufficiently different from that of the other samples that we have questioned whether this behavior can be satisfactorily explained within a conventional size-effect theory. In all samples at high fields, the magnitude of the linear term does not appear to be affected by the wire diameter.

IV. DISCUSSION

A. Macroscopic Theories of Linear Magnetoresistance

The first theories that were applied to the linear magnetoresistance of potassium were motivated originally by experiments in semiconductors and considered effects such as sample geometry and inhomogeneities. Lippmann and Kuhrt[18] solved the boundary-value problem of a rectangular conducting plate in a transverse magnetic field with current flowing between two electrodes separated by a distance a extending along opposite edges of length b. For a large magnetic field and a finite aspect ratio b/a this geometry results in a linear magnetoresistance. As the Hall angle $\theta_H = \tan^{-1}(\omega_c\tau)$ approaches $\frac{1}{2}\pi$, the Kohler slope becomes equal to the aspect ratio.

Babiskin and Siebenmann[3] have reported reasonable quantitative agreement with the theory of Lippmann and Kuhrt. The aspect ratios of their wire samples ranged from $b/a = 0.07\times10^{-2}$ to 4×10^{-2} in specimens having a purity range corresponding to $\omega_c\tau$ values ranging from 30 to 300 at 90 kG. However, in our experiments we have found no such agreement. To within 15%, all of our 1.5-mm-diam wire samples had the same aspect ratio $b/a = 0.2\times10^{-2}$. Among these samples, the Kohler slope varied by almost a factor of 30 for only a factor of 3 range in $\omega_c\tau$ at 50 kG (from 30 to 90). The reasonable agreement between the Kohler slopes of our long wires and the matchstick-sized samples of Lass ($b/a \approx 5\times10^{-2}$) also seems to rule out a strictly geometrical effect. Finally, we note that the Kohler slopes observed for our long-wire samples are of comparable magnitude to those observed in completely different sample geometries such as the thin discs used in the helicon measurements of Penz and Bowers and the spheres employed by Lass in the induced torque experiments.

Herring[19] has predicted a linear magnetoresistance due to isotropic bulk inhomogeneities. A Kohler slope of 10^{-2} would require a variation of 10% in the carrier density within the sample. As Penz and Bowers[1] have pointed out already, this is an unreasonably large variation for a metal.

With a respect to both the geometrical and inhomogeneity arguments, we should also comment on Babiskin and Siebenmann's observed correlation between the slope of the linear term and the number of contraction voids on the surface of the sample. We do not dispute the existence of such a correlation, but we do question the extent to which this observation can be used to explain the linear term in other experiments. The results of both Lass and Penz and Bowers on good single crystals make it difficult to believe that sample voids are the sole cause of the linear term. Our sample treatments such as annealing and freezing in oil not only affected the high-field linear term but changed the low-field magnetoresistance as well. This suggests that the analysis of Babiskin and Siebenmann in which they separate the magnetoresistance into an intrinsic saturating component and a linear term attributed to sample voids is premature.

B. Intrinsic Theories of Linear Magnetoresistance

It is well known that the theory of Lifshitz, Azbel, and Kaganov (LAK)[20] predicts high-field saturation of the transverse magnetoresistance of metals such as potassium which are uncompensated and allow only closed electron orbits. LAK expand the solution of the Boltzmann equation in inverse powers of the magnetic field H. In neglecting interband transitions, the band structure of the metal enters the theory only through the topology of the electron-cyclotron orbits. The type of orbit determines the leading term in the $1/H$ expansion of each component of the magnetoconductivity tensor. It is implicitly assumed that the convergence of the series in $1/H$ is sufficiently rapid that only the leading term is significant. No detailed assumptions on the form of the collision operator are made except that it be independent of H.

To seek the origin of a linear magnetoresistance which is consistent with the LAK theory, each assumption as it applies to potassium must be scrutinized carefully. One such assumption is the presumed absence of open and holelike orbits on the Fermi surface of potassium. Both Reitz and Overhauser[21] and more recently O'Keefe and Goddard[22] have proposed the existence of small energy gaps which can give rise to open and holelike orbits. The physical origin of the gaps is quite different in these two models. Reitz and Overhauser attribute the gap to the existence of either a charge density or spin density wave in the metal while O'Keefe and Goddard obtain a different band structure for the alkalis by applying a new antisymmetrizing scheme to the electron wave functions. The magnetoconductivity is calculated in both models by treating the magnetic breakdown of the gaps by the method of Falicov and Sievert.[23] Each can produce a linear term up to perhaps $\omega_c\tau \approx 150$. Above this value, the magnetoresistance begins to saturate as the electrons tunnel through the gaps and return to free-electron-like closed orbits. Both models also predict a decrease in the high-field Hall coefficient as the holelike orbits disappear as a result of the breakdown of the gaps.

Another theoretical approach to the linear magnetoresistance has been to leave the spherical Fermi surface intact and to investigate the effect of extremely anisotropic scattering of the electrons. The rate of convergence of the LAK power series in $1/H$ is measured by the dimensionless parameter $1/\omega_c\bar{\tau}$, where $\bar{\tau}$ is some relaxation time averaged over an electron orbit.[20,24] If electron scattering is extremely anisotropic, $\bar{\tau}$ may not be a sufficiently well-defined quantity to permit rapid convergence of the series at low fields.

Young[25] has constructed a model which illustrates this fact. He considers the effect of electron-phonon umklapp scattering which takes place at regions of the Fermi sphere which approach most closely the first-Brillouin-zone boundary. These localized scattering centers are referred to as "hot spots." Young solves the Boltzmann equation by assuming two scattering rates, one for umklapp scattering within the hot spots and another for background impurity scattering over the entire Fermi sphere. The two free parameters of the model are the umklapp scattering rate per unit solid angle, B, and the solid angle of the hot spot, Ω_0. The difficulty with this model arises from the extreme values of these parameters which are needed to obtain agreement with experiment. For a Kohler slope of $\sim 10^{-3}$, the umklapp scattering rate $B\Omega_0$ must be almost 10^4 times larger than that from impurities. In addition the hot-spot size must be exceedingly small, $\Omega_0 \sim 10^{-5}$ sr. Less extreme values of B and Ω_0 result in saturation of the magnetoresistance in the field range which has been investigated experimentally. As with the band gap proposals, Young's model predicts a decrease in the high-field Hall coefficient.

In a sense, the source of the linear term in all of these theories is the same. It is simply Bragg reflection at a well-localized scattering center in k space, be it a band gap or an umklapp hot spot. In each case there is a band of electrons on the Fermi sphere which contributes a well-defined flux onto the scattering center proportional to the cyclotron frequency ω_c, and hence a large angle scattering rate proportional to the applied magnetic field. A resistive mechanism results when there is a net amount of Bragg scattering against the flow of current in the sample.

C. Comparison of Intrinsic Theories with Experiment

Probably the most distinguishing feature of each of the intrinsic theories is the anisotropy that each predicts in the magnetoresistance. None of the single-crystal experiments that have been performed can support a very strong anisotropy of the linear term. Penz and Bowers found such a large range of Kohler slopes among samples of the same crystal orientation that no definite conclusions could be drawn. The rotation studies on unoriented single crystals by Lass produced no anisotropy other than that which could be accounted for by departures from specimen sphericity.

The impurity dependence of the Kohler slope which we observe [Fig. 9(b)] might conceivably be consistent with a magnetic breakdown interpretation. One would expect the Kohler slope to decrease at high purities as the breakdown parameter $\omega_0\tau$ and the number of attempts by an electron at a band gap increase.[23] Balcombe and Parker[26] have calculated similar deviations from Kohler's rule for their magnetic breakdown model of aluminum. However, the magnetoresistance predicted by O'Keefe and Goddard does not appear to be very sensitive to sample purity for large $\tau\,(\vec{H}\,\|\,[001])$.[27] The reason may be that, in contrast to the case of aluminum, electrons with a relatively wide range of $k_z(\vec{H}\,\|\,z)$ and orbit diameter are participating in the breakdown process.

The decrease which we observe in the Kohler slope between intermediate- and low-purity samples could be explained by an extremely short mean free path in the alloys. This ensures that an electron always impurity scatters before Bragg reflecting at a band gap ($\omega_c\tau < 1$ for fields in the breakdown region). The O'Keefe-Goddard calculation does bear this out in that the slope of the linear portion decreases for small $\tau\,(\vec{H}\,\|\,[001])$.[27]

One of the biggest problems with the intrinsic theories, both umklapp and band gap, is that they predict the onset of saturation at fields which are too low to be compatible with experimental observations. In all experiments on both single and polycrystals, there has been no indication of saturation at the highest attainable fields. Indeed, Penz and Bowers have observed the linear term for $\omega_c\tau$ as high as 300. For the work reported here, the deviations from linearity above the knee are at most $\frac{1}{2}\%$ of the zero-field resistivity for $\omega_c\tau$ values as large as 150.

In particular, the band-gap models require an energy gap of about 0.06 eV in order to delay saturation to a sufficiently high field. A gap this large must be reconciled with de Haas–van Alphen (dHvA)[28] measurements which show the Fermi surface to be spherical to about 1 part in 10^3. Reitz and Overhauser[21] have offered some possible explanations for this apparent inconsistency. As they point out, it might be useful to extend dHvA measurements to lower fields. The experiment of Shoenberg and Stiles[27] was done at a fixed field of about 50 kG and therefore could be insensitive to changes in electron orbits due to magnetic breakdown. A determination of the field dependence of the dHvA amplitude in the region of H_0, the onset field of the linear term in the magnetoresistance, might be especially useful. However, it should be mentioned that swept-field magnetoacoustic studies of the Fermi surface[29] in the range 0–15 kG agree closely with the dHvA results. Reitz and Overhauser have also suggested that a decrease in dHvA signal amplitude due to internal defects and strain in the specimens might explain why no evidence for small band gaps has been found. In spite of these considerations, Fermi-surface studies do seem to indicate that at most only an extremely small fraction of the total number of conduction electrons could be participating in any magnetic breakdown process.

While the intrinsic theories have difficulty in producing a linear magnetoresistance of sufficient magnitude at high fields, they all yield a decrease in the high-field Hall coefficient; this is qualitatively the same behavior as observed experimentally by Penz.[30] The challenge to future theories would seem to be to retain a linear term in the magnetoresistance to higher fields and yet give the correct field dependence of the Hall coefficient. In this regard, experiments which could relate variations in the observed onset field and Kohler slope to corresponding changes in the Hall coefficient would be very helpful.

Another difficulty with models based on either magnetic breakdown or umklapp scattering is that, as proposed, they are only applicable to the transverse magnetoresistance. The comparable magnitude of the Kohler slope for longitudinal and transverse magnetoresistance as observed by both Jones and Lass suggests that the same mechanism may be responsible for the linear term in these two different configurations. Lass has presented some evidence that the linear term is even larger in the longitudinal case; successive measurements made on the same sample without altering the probes showed the Kohler slope of the longitudinal magnetoresistance to be about 30% higher than that of the transverse.

Besides extreme values of parameters needed to fit the observed Kohler slopes, the electron-phonon umklapp model has difficulty in explaining the absence of a temperature dependence of the linear term. If the linear magnetoresistance were due to electron-phonon umklapp scattering, then at a sufficiently low temperature where umklapp processes are frozen out, the magnetoresistance should saturate. Theoretical[31] and recent experimental[32] studies of the temperature dependence of the zero-field resistivity show that the contribution of electron-phonon umklapp scattering to the ideal resistivity is negligible below 1.5 K. This suggests that the solid angle Ω_0 of the hot spots should have decreased enough in the temperature range of our experiment to give an observable reduction in the Kohler slope. Our results are inconsistent with the prediction of any decrease in the Kohler slope with decreasing temperature.

Babiskin and Siebenmann have suggested that electron-phonon umklapp scattering could contribute to the nonlinear low-field magnetoresistance. The fact that we observe both H_0 and $(\Delta\rho/\rho_0)_{H_0}$ to increase with temperature is consistent with this proposal. As the temperature is increased a low-field magnetoresistance due to umklapp scattering could dominate the temperature-independent linear term to increasingly higher fields. Preliminary measurements have shown a large increase in the low-field magnetoresistance for temperatures above 4.2 K.[32]

A question which can be posed for all of the intrinsic models is the extent to which they would be affected by a large background of small-angle scattering from phonons or extended defects. The bands of electrons incident upon an energy gap or hot spot could be fed or depleted by electrons undergoing small-angle collisions. This possibility is suggested by studies of Pippard[33] on the magnetoresistance of metals such as copper and aluminum which have more anisotropic Fermi surfaces than potassium. In copper, for example, a small-angle collision can lead to Bragg reflection if it takes the electron into a neck orbit passing through the zone boundary.

In potassium, the effect of small-angle anisotropic scattering from dislocations might be enough to obscure the orientational dependence of the Kohler slope that is predicted by the intrinsic theories. However, it seems unlikely that small-angle scattering can be the sole cause of the linear term. The Kohler slope decreases significantly for higher-purity samples where one expects small-angle collisions to play a more important role. In fact, at the highest purities, the Kohler slope is close to the value for alloy samples where isotropic collisions with impurity atoms should be the dominant scattering mechanism. Also, the temperature independence of the Kohler slope does not link it to small-angle normal-phonon scattering.

On the other hand, small-angle dislocation or normal-phonon scattering might help to explain the observed deviations from linearity at low fields. We have already seen how the low-field magnetoresistance invariably decreased with both annealing and temperature. We would not expect annealing to significantly affect the amount of large-angle scattering of electrons except in those cases where the RRR and the negative magnetoresistance of the size effect significantly increased. The indication that internal defects enhance the temperature dependence of the low-field magnetoresistance might possibly be explained in terms of an umklapp "hot-spot" model. Small-angle dislocation scattering could carry an electron into an orbit passing through a hot spot, thus effectively widening the bands of electrons on the Fermi sphere that are engaged in umklapp scattering.

V. CONCLUSIONS

Our measurements of the transverse magnetoresistance of polycrystalline samples under a variety of experimental conditions have demonstrated that at least some of the previously conflicting results in potassium have been due to differences in sample preparation and handling. The appearance of the quasisaturation phenomenon in well-annealed intermediate-purity samples and the temperature dependence of the low-field magnetoresistance in the purest samples suggest that the nonlinear behavior at low fields is intrinsic to the sample; that is, it is not to be associated with errors in measurement. A high-field linear magnetoresistance was present in all samples under all conditions, and is therefore also thought to be a property of the sample.

We are unable to characterize our polycrystalline wire samples well enough for our data to be used quantitatively in constructing a theory of the magnetoresistance of potassium. Rather, the patterns which we observe in our well-annealed polycrystalline samples suggest the following constraints on a theory of the magnetoresistance of potassium: (i) It must be able to produce a linear magnetoresistance which persists at least as high as $\omega_c\tau \approx 150$; (ii) the Kohler slope must be nearly independent of temperature below 4.2 K; (iii) the theory must give an impurity dependence of the linear term (for polycrystalline samples) which is consistent with that in Fig. 9(b); (iv) the pressure derivative of the magnetoresistance and the zero-field resistivity should be of opposite sign; (v) it must allow for a decrease in the low-field magnetoresistance with both annealing and a decrease in temperature; (vi) the theory must give the correct field dependence of the Hall coefficient as well as the linear term in the magnetoresistance at high fields.

It would be difficult to conclude that any of the existing theories adequately describe the observed magnetoresistance. It is not clear how the patterns in the data could support the macroscopic theories which have been used to explain the linear term. As for the intrinsic theories, the temperature independence of the Kohler slope is probably the most compelling evidence against electron-phonon umklapp scattering being responsible for the linear term, although umklapp scattering could be contributing to the temperature-dependent nonlinear magnetoresistance at low fields. To produce a linear magnetoresistance at $\omega_c\tau$ of 150, the band-gap theories generally require energy gaps which are too large to be reconciled easily with the dHvA results on potassium.

Despite these discrepancies, it is unfortunate that our polycrystalline samples do not permit a more complete comparison with the intrinsic theories. In view of the patterns we have observed in the magnetoresistance of annealed polycrystalline samples, measurements on well-annealed single crystals may now prove more fruitful. Anisotropy in the observed magnetoresistance and Hall coefficient might well provide the most decisive test of an intrinsic linear magnetoresistance of potassium.

Note added in proof. Since submitting this manuscript, we have extended our magnetoresistance measurements to two higher-purity annealed wire samples of RRR = 5450 and 6090. These samples have Kohler slopes of 0.41×10^{-2} and 0.24×10^{-2} (rad^{-1}), respectively. The shape of the magnetoresistance curves at low fields is qualitatively similar to sample KX13 in Fig. 4(a). These new results are consistent with the pattern in the impurity dependence of the Kohler slope shown in Fig. 9(b). The Kohler slope remains at about the same value of 0.4×10^{-2} (or less, depending upon annealing) for RRR > 2800. There is no evidence that the Kohler slope increases in the highest-purity samples.

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