High-field transverse and longitudinal thermal magnetoresistances of potassium

P. J. Tausch,* R. S. Newrock, and W. Mitchel
Physics Department, University of Cincinnati, Cincinnati, Ohio 45221
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We report the results of extensive thermal magnetoresistance measurements on polycrystalline potassium specimens. The specimens had residual resistance ratios ranging from 2100 to 7300. Measurements were made between 3 and 9 K, for magnetic fields up to 9.5 T. No sign of saturation was observed. The previously observed quadratic term persists to the highest fields, completely dominating the linear term. The various possible sources of error are carefully analyzed and discussed and we conclude that the quadratic term is not due to probe effects or measurement artifacts. Using previously determined values of the lattice thermal conductivity k_g, we correct the data for the effects of lattice heat conduction. The corrected thermal magnetoresistance has the same form as the uncorrected: $W(H,T)T = W(O,T)T + AH + BH^2$. Values of A and B, corrected and uncorrected for k_g, are presented. $A = A_0 + A_1T^3$, with A_0 strongly purity dependent and A_1 almost independent of purity. B is also almost independent of purity and is a monotonically decreasing function of T. The thermal Kohler slope is compared to the electrical Kohler slope and is found to be nearly four times larger for specimens of similar purity. A speculation concerning the role of deviations from Matthiesson's rule on the Kohler slopes is presented. Kohler's rule is obeyed. The Wiedemann-Franz law is not obeyed, even if the quadratic term is ignored.

I. INTRODUCTION

It is now well known that the magnetotransport coefficients of potassium differ strongly from the predictions of the semiclassical theory of transport in metals. None of the predicted magnetic field dependences of the diagonal elements of the electrical and thermal magnetoresistivity tensors are borne out by experiment. This paper presents data from a series of measurements which extend our previous work[1,2] on the transverse thermal magnetoresistivity to much higher fields. The magnetic field ranged from zero to above 9 T, the temperature from 2 to 8 K, and the residual resistance ratios (RRR) of the specimens from 1000 and 7300. We also report several preliminary measurements of the longitudinal thermal magnetoresistivity. We show how much of the intermixing among the various thermal-conductivity-tensor elements can be sorted out with the help of the high-field measurements. Some of the implications of the new data concerning a viable magnetoresistance theory for the simple metals are also discussed.

We note that when the measurements presented here are included, all of the pure electric and thermal magnetotransport coefficients have been measured. Thus it is now possible to make extensive comparisons between the data and the proposed theories, both extrinsic and intrinsic, and to speculate about the possible origins of the magnetoresistance anomalies. These comparisons will be presented in a future article.

II. EXPERIMENTAL DETAILS

Many of the details of the experimental procedures have been previously published;[1,3-5] in this section the important points are outlined.

A. Specimens

It is well known that specimen to specimen variations are of considerable importance in transport measurements in potassium. We therefore describe our sample fabrication procedures in some detail. All of the specimens used in this work were fabricated from 99.99% pure potassium supplied in argon-filled glass ampules.[6] At the beginning of the fabrication process the end of the ampule is broken off and the ampule is heated under vacuum to a temperature just above the melting point. The potassium is allowed to flow into glass molds designed for single-crystal growth and easy ejection of the boules. The molds are carefully cleaned and coated with dry paraffin oil before use. The potassium is held above the melting point under a vacuum of about 10^{-5} Torr for 24–48 h, and then allowed to cool slowly over a period of 6–8 h. After ejection from the molds the boule is cleaned, etched (secondary butyl alcohol in xylene, 2% by volume), and stored under dry paraffin oil in an evacuated desiccator.

The specimens are polycrystalline and were formed in a stainless-steel sample press using the following procedure: the oxide layer is removed from a portion of the boule, and the piece is placed between the oil-coated plates of the

sample press and pressed to the desired thickness. A special jig is used to slice the flat plate formed into several specimens, which are etched, placed in an evacuated desiccator under dry paraffin oil, and allowed to anneal at room temperature for a minimum of 48 h. After annealing, the samples are generally found to consist of two to three large crystallites. (In our previous work[1] we used specimens that had been extruded or were single crystals, as well as those prepared with the press. The thermal-magnetoresistance results did not depend significantly on the method of specimen preparation nor upon the specimen's being a single crystal; for this work we therefore used only pressed polycrystals.)

The geometrical parameters of the specimens are listed in Table I. The error in the geometrical factors is estimated to be 4% for the width and 2% for the thickness. The average distance between the measuring thermometers was slightly different in different specimens, about 1.5 cm with an uncertainty of about 5%.

After annealing, the specimens are carefully mounted following the procedures outlined in Refs. 1 and 5. The specimen is supported by the measuring thermometers, with thermal contact to the thermometers made via a drop of Dow-Corning 200 Fluid.[7] A small "clamp" and a drop of DC-200 fluid at the platform end ensures reasonable thermal contact to the helium bath. A tweezerlike heater,[3] attached to the other end (also with a drop of DC-200 fluid), provides the heat current. After mounting, the specimen is cooled slowly (over a 45-min period) to liquid-nitrogen temperatures.

B. Measurement apparatus and electronics

Standard linear heat flow techniques are employed in these experiments. The measuring thermometers are germanium resistance thermometers,[8] previously calibrated against temperature, and calibrated during each experiment versus magnetic field.

The magnetic field is supplied by a 1.5-in.-bore superconducting solenoid, nominally rated at 8 T at 4.2 K, but capable of 10 T for $T \lesssim 2.4$ K. The field of the solenoid was previously calibrated versus the magnet current using a rotating coil gaussmeter[9]; the calibration is accurate to about 0.1%.

C. Data acquisition

Before measuring the thermal magnetoresistance we measure the zero-field thermal resistance of each specimen between 2 and 9 K to aid us in characterizing the specimens. The zero-field thermal resistance of potassium, $W(0,T)$, is known[3] to have a simple power-law temperature dependence to temperatures in excess of 10 K:

$$W(0,T) = A_0/T + B_0T^2 . \tag{1}$$

The values of the coefficients A_0 and B_0 are listed in Table I for each specimen. Employing our previous procedures,[1,3] we determine the residual-resistivity ratio r_R from the coefficient A_0, using Eq. (1) and the Weidemann-Franz law,

$$r_R = \frac{\rho(0,273)}{L_0[W(0,T)T]_{T=0}} = \frac{\rho(0,273)}{A_0L_0} ,$$

where L_0 is the Lorenz number. Comparisons with electrical determinations of the RRR using the method of Bean *et al.*[10] have shown that this method provides values of RRR that are accurate to 5% or better.[1] Table I shows the RRR for our specimens. For $\rho(0,273)$ we used 7.16×10^{-8} Ω m.[11]

After the zero-field data are obtained the thermal magnetoresistance is measured using the procedures outlines in Refs. 1 and 5. Temperature is regulated with an active-bridge temperature controller,[12] which uses a strontium-titanate capacitance thermometer[13] as a temperature sensor. Both the capacitance and dissipation of these sensors are independent of magnetic field,[14] enabling the controller to maintain the specimen temperature to within a few mK as the magnetic field is changed. To ensure that the magnitude of the magnetic fields in the normal and reverse directions

TABLE I. Geometric and zero-magnetic-field characterization of the specimens.

Sample	A_0 [a] $= (WT)_{T=0}$ (10^{-4} m K^2/W)	B_0 [a] (10^{-5} m/W K)	RRR [b]	Width (mm)	Thickness (mm)
KHF-1	4.8	1.75	6100	3.0	1.0
KHF-2	7.5	1.68	3900	3.0	1.0
KHF-5	10	1.86	2900	8.0	1.2
KHF-7	14	2.13	2090	8.0	1.0
KHF-8	4.0	1.75	7300	8.0	1.3

[a] The coefficients A_0 and B_0 are obtained by fitting the data to Eq. (1) described in the text.
[b] The residual resistivity ratio as determined from the Wiedemann-Franz law; see text.

are matched to 0.05% or better a sensitive bismuth magnetoresistor[15] is employed. During reverse field runs the magnetic field is adjusted until the resistance of the bismuth probe is matched to its value in the normal field direction. This procedure is necessary for fields up to 2 T. Above that the hysteresis of the magnet is negligible, and to insure equal normal and reverse fields it is sufficient to match magnet currents.

III. RESULTS

In order to keep the experimental results and data separate from the analysis and interpretation this section presents the actual experimental results together with a few brief comments and observations. Analysis and interpretation of the data are in the next several sections.

The magnetic field and temperature dependence of the transverse thermal magnetoresistivity of potassium are shown in Figs. 1 and 2 for some of the data on a high-purity specimen, KHF-8. Similar data for a second specimen, KHF-1, have been published elsewhere[16] and the data for the other specimens used in this study are available from the authors. In these figures the thermal magnetoresistivity times temperature, $W(H,T)T$, is plotted as a function of the applied magnetic field. $W(H,T)T$ is the thermal analog of the electrical magnetoresistivity.

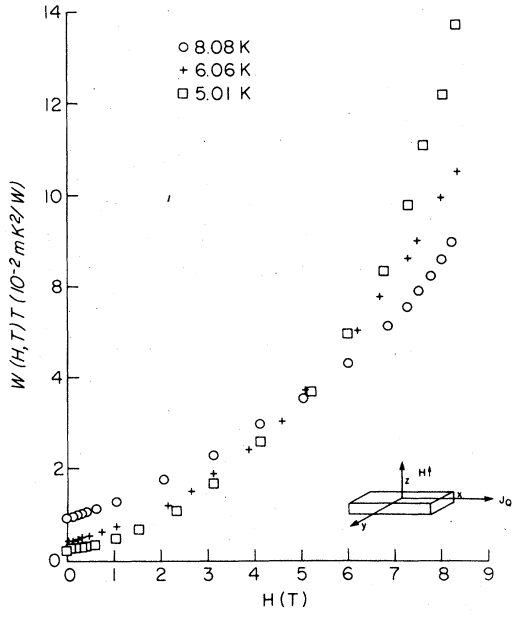

FIG. 1. Transverse thermal magnetoresistance of potassium times the temperature as a function of the applied magnetic field; specimens KHF-8, for high-temperature data as indicated. Note the absence of saturation and the crossing behavior.

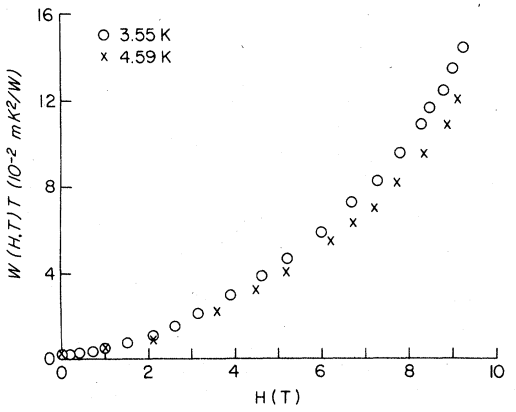

FIG. 2. Transverse thermal magnetoresistance of potassium times the temperature as a function of the applied magnetic field; specimen KHF-8, two low temperatures. Note that the curves cross as the field increases.

These are several important features to be noted in these data, some of which were already evident from our earlier work at lower fields.[1]

(i) The relative change in the thermal magnetoresistivity with magnetic field is extremely large, and is so for all purities. The relative change is 75–300 times greater than in the electrical case. For specimen KHF-5, for example, $\Delta W(9\mathrm{T}, 3.4\mathrm{K})/W(0, 3.4) \cong 140$, whereas for a specimen of similar purity (e.g., Taub *et al.*,[17] KX-17) $\Delta\rho(0, 3.4)/\rho(9\mathrm{T}, 3.4) \approx 0.8$. As a further illustration of these differences Fig. 3 displays

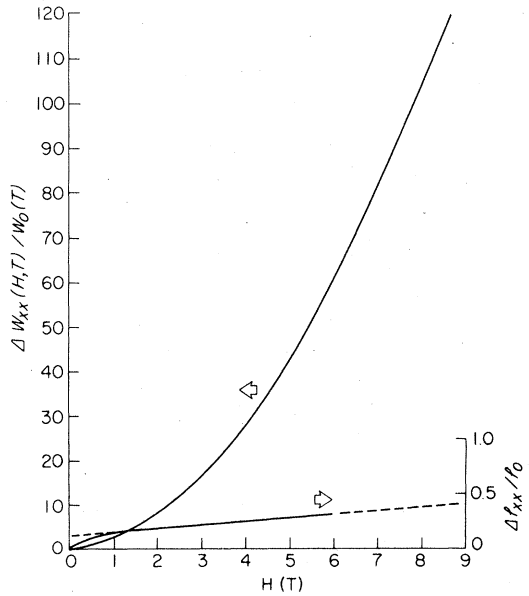

FIG. 3. Comparison of the reduced transverse electrical $(\Delta\rho/\rho_0)$ and thermal $(\Delta W/W_0)$ magnetoresistivities for specimens of similar purity.

$\Delta\rho(H,T)/\rho_0$ and $\Delta W(H,T)T/W_0(T)$ versus magnetic field for specimens of similar purity.

(ii) The previously observed[1,2,18] quadratic field dependence clearly dominates the thermal magnetoresistance in strong fields. We find no evidence of the predicted saturation[19] up to magnetic fields greater than 9 T ($\omega_c\tau > 300$). The transverse electrical magnetoresistivity is, of course, linear in field over this range,[17] and also shows no sign of saturation.

(iii) The transverse thermal magnetoresistance is strongly temperature dependent. This is in striking contrast to the transverse electrical magnetoresistivity, which has a relatively weak temperature dependence.

(iv) The purity dependence of the thermal magnetoresistance is relatively weak. This is again in direct contrast to the electrical case, where the purity (or state of strain) of the specimen, as measured by RRR, is known to have a large effect on the Kohler slope,[17] defined by $\Delta\rho/\rho = S\omega_c\tau$.

(v) The temperature dependence of the thermal magnetoresistivity is complicated. The relative change in $W(H,T)T$ decreases as the temperature increases. It is shown below that most of the temperature dependence is in the quadratic term. From Fig. 2, it is clear that a curve of $W(H,T)T$ vs H at a lower temperature will eventually cross a higher-temperature curve. Thus at high constant values of the magnetic field the thermal resistance *increases* as the temperature *decreases*.

(vi) Several points mentioned in Ref. 1 are worth repeating:

(a) The thermal magnetoresistance increases monotonically with the applied magnetic field. There are no signs of knees, quasisaturation, nor negative magnetoresistance, effects which are observed frequently in the electrical magnetoresistivity.[17,20]

(b) Using a mean scattering time τ_{el} determined from simple free-electron models ($\tau_{el} = m/ne^2\rho$), we can show that Kohler's rule does not hold, that is, $W(H,T)T$ does not scale as a function of $\omega_c\tau_{el}$.

(c) The high-field Lorenz ratio,[21] $L = \rho(H)/W(H,T)T$, is field dependent, and decreases with increasing field. Since the largest changes in the thermal magnetoresistivity with field occur at low temperatures, the deviation from the Wiedemann-Franz ratio increases as the temperature is lowered, or as the field increases. We note that the low-temperature region is precisely the region where the Wiedemann-Franz law is expected to be valid since at low temperatures elastic impurity scattering should be the dominant electron scattering mechanism.

Several preliminary experiments have been performed on the longitudinal thermal magnetoresistivity. The results of one of these experiments are shown in Fig. 4. There are two preliminary observations we may make: (i) There is no sign of a quadratic term. The longitudinal thermal magnetoresistivity $W_{zz}(H,T)T$ is linear over the entire field range. (ii) The Kohler slope is roughly the same as that of the linear portion of the transverse thermal magnetoresistivity (see Secs. IV and V).

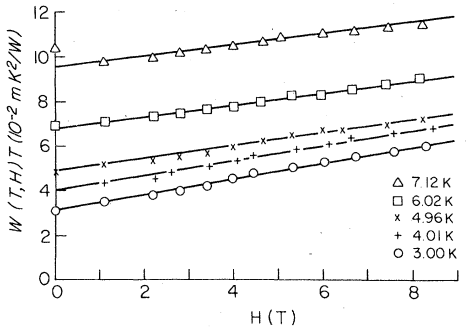

FIG. 4. Longitudinal thermal magnetoresistance of potassium times the temperature as a function of the applied magnetic field for several temperatures.

IV. DATA ANALYSIS

The previous lower-field measurements of the thermal magnetoresistivity of potassium[1,18] showed that the data can be fit to a polynomial of the form

$$W(H,T)T = W(0,T)T + AH + BH^2, \qquad (2)$$

where the coefficients A and B are temperature dependent, A is purity dependent, and B has no discernible purity dependence. This equation also provides an excellent fit to the data taken at the much higher fields used in this work; no higher-order terms are needed. The quality of the fit may be seen from Fig. 5, which shows a plot of

$$\Delta W(H,T)/H = [W(H,T) - W(0,T)]/H$$

versus H at several temperatures for specimen KHF-5. The lines are the best fits to the data. The coefficient A in Eq. (2) is obtained from the zero-field intercept and the coefficient B from the slope. Note that the curve is linear up to the highest fields. All of the specimens were analyzed in this manner. The results are displayed in Table II and discussed below.

Before discussing the data we must investigate the possibility that the quadratic term is spurious. There is a considerable body of evidence showing that experimental artifacts and probe effects are not the cause of the linear term and we will not discuss this here. (Ref. 17 takes up this point in

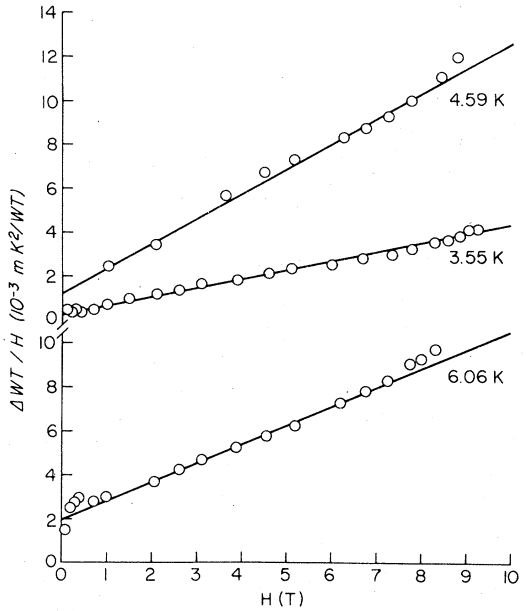

FIG. 5. Change in the thermal resistivity with magnetic field divided by the field as a function of the applied field for specimen KHF-8 at several representative temperatures. The slope yields the coefficient of the quadratic term and the intercept that of the linear term in the transverse thermal magnetoresistivity [Eqs. (2) and (6)].

detail.) There are a number of possibilities to be considered: (i) are the specimens well into the high-field limit; indeed, what is the high-field limit for thermal measurements (ii) is the nonzero lattice thermal conductivity responsible for the quadratic term and (iii) are there probe effects or other measurement artifacts?

A. The high-field limit

It may be shown[22] from quite general considerations that both of the magnetoresistivities of a metal are quadratic in the applied magnetic field for $\omega_c\tau \ll 1$. In the electrical case, $\omega_c\tau = 1$ for fields of the order of 0.02–0.03 T for very pure potassium, and the low-field quadratic term is not observed in the electrical magnetoresistivity measurements. Because of the important role played by small-angle inelastic scattering in the thermal resistivity the thermal scattering time will differ greatly from the electrical scattering time, perhaps by factors of 10 or 100, and it is possible that the observed behavior of $W(H,T)$ is due to the fact that the high-field limit is not reached. Thus we must determine the mean time between scattering events important in thermal conduction. In a previous article[5] we showed that an operational definition of the thermal scattering time can be made, namely, $\omega_c\tau_{\rm th} = \nabla_y T/\nabla_x T$. The scattering times obtained in this manner, from simultaneous measurements of $\nabla_y T$ and $\nabla_x T$, have the correct temperature dependence and a magnitude in good agreement with theory. $\tau_{\rm th}$ was somewhat larger than predicted, a reasonable result as the theory gives the total number of scattering events per unit time, but includes no measure of the relative effectiveness that each scattering event has in degrading the thermal current. From the data presented in Ref. 5 we may determine the magnetic field H_1 at which $\omega_c\tau_{\rm th} = 1$. These values are listed in Table II. It may be seen that it was possible to obtain magnetic fields well in excess of H_1, i.e., $\omega_c\tau_{\rm th} \gg 1$ for all temperatures and purities used in this work. Only those data points for which $\omega_c\tau_{\rm th} \geq 5$ were used in determining the values of the coefficients A and B. Thus we conclude that the quadratic term is not a remnant of the low-field limit.

B. Effects of the lattice conductivity

It has been proposed that the quadratic term may be due to the effects of the lattice thermal conductivity.[18] If a tensor representing the lattice conductivity $\vec{\vec{K}}_g = k_g\vec{\vec{1}}$ is added to the electronic thermal conductivity tensor $\vec{\vec{K}}_e$, it is simple to show that[5,23]

$$W^M_{xx}(H,T)T = W^e_{xx}(H,T)T\left[\frac{1+k_gW^e_{xx}+k_g(W^e_{xy})^2/W^e_{xx}}{(1+k_gW^e_{xx})^2+(k_gW^e_{xy})^2}\right], \tag{3}$$

and

$$W^M_{xy}(H,T)T = TR^M_{\rm RL}H = W^e_{xy}(H,T)T \times[(1+k_gW^e_{xx})^2+(k_gW^e_{xy})^2]^{-1}, \tag{4}$$

where the superscript M refers to the measured transport coefficient, the superscript e refers to the transport coefficient that would be measured if the lattice conduction were zero, and $R_{\rm RL}$ is is the Right-Leduc (thermal Hall) coefficient.

In most of the simple metals $k_gW^e_{xx}$ and $k_gW^e_{xy}$ are very small compared to unity, and Eq. (3) can be approximated as

$$W^M_{xx}(H,T)T = W^e_{xx}(H,T)T + k_gT[H^2/(L_0Tne)^2], \tag{5}$$

where we have assumed $W^e_{xy} = H/L_0Tne$. It is therefore tempting to ascribe the quadratic magnetic field dependence of the thermal magnetoresistivity of potassium to lattice conduction. If Eq. (5) is fit to the thermal-magnetoresistivity data a very large lattice conductivity (five to nine times larger than expected) with an anomalous temperature dependence[18] is found. Figure 6 shows a plot of the average lattice thermal con-

TABLE II. Summary of important experimental data.

	T (K)	A $10^{-3}\frac{K^2 m}{WT}$	B $10^{-2}\frac{K^2 m}{WT^2}$	A^e $10^{-3}\frac{K^2 m}{WT}$	B^e $10^{-2}\frac{K^2 m}{WT^2}$	A_0^e $10^{-3}\frac{K^2 m}{WT}$	A_1^e $10^{-5}\frac{m}{WTK}$	$10^2 S_T^0$	$10^2 S_E$	H_1 (T)
KHF-1	2.00	0.15	0.127	0.15	0.126	0.1	0.68	0.58	0.25	0.036
	3.01	0.35	0.125	0.27	0.126					0.059
	4.01	1.1	0.104	0.88	0.104					0.106
	5.00	1.05	0.083	1.00	0.081					0.181
	6.04	1.65	0.058	1.63	0.056					0.456
	8.01	2.55	0.028	2.59	0.019					0.666
KHF-2	2.00	0.2	0.136	0.16	0.137	0.05	1.21	0.29	0.20	0.050
	3.00	0.4	0.136	0.24	0.128					0.075
	3.74	0.7	0.116	0.67	0.110					0.105
	5.00	1.55	0.087	1.45	0.083					0.190
	6.01	2.8	0.027	2.58	0.024					0.310
	8.06	2.7	0.031	2.82	0.018					0.680
KHF-5	3.44	1.0	0.134	0.96	0.136	0.96	1.36	2.8	0.6	0.105
	4.59	1.75	0.104	1.51	0.104					0.165
	6.08	3.10	0.044	2.74	0.047					0.325
	6.88	3.25	0.037	3.37	0.025					0.430
KHF-7	3.00	1.30	0.174	1.27	0.168	1.06	0.86	5.8	1.5	0.109
	3.44	2.25	0.135	2.33	0.132					0.145
	4.00	1.80	0.149	1.68	0.149					0.156
	4.58	2.0	0.127	1.92	0.126					0.194
	5.01	2.5	0.108	2.1	0.099					0.231
	6.07	3.0	0.071	2.97	0.065					0.346
	6.87	4.0	0.044	3.98	0.043					0.486
	8.05	5.8	0.026	5.3	0.023					0.731
KHF-8	2.55	0.4	0.148	0.76	0.148	0.4	0.63	2.2	0.3	0.043
	3.55	0.8	0.149	0.71	0.150					0.079
	4.59	1.10	0.119	1.06	0.112					0.144
	5.01	1.15	0.117	1.20	0.107					0.182
	6.06	1.85	0.085	1.82	0.081					0.303
	7.07	2.55	0.051	2.55	0.042					0.468
	8.08	3.15	0.041	3.23	0.034					0.681

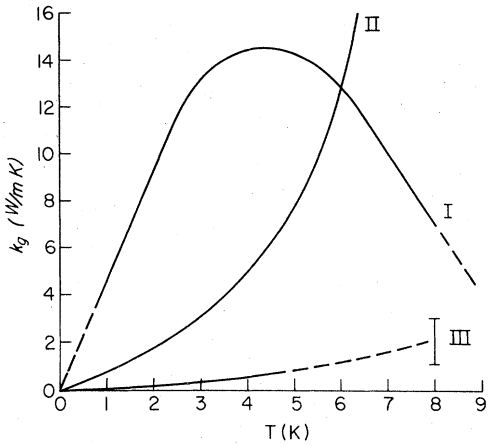

FIG. 6. Lattice thermal conductivity of potassium: curve I, as determined from the quadratic term in the thermal magnetoresistivity (see text); curve II, theoretical calculation of Ekin; curve III, as determined from simultaneous measurements of $W_{xx}(H,T)$ and $W_{xy}(H,T)$ (this work and Ref. 5).

ductivity obtained in this manner from several specimens (curve I). In addition the theoretical calculation of Ekin[24] (curve II) is shown. It is clear that the thermal conductivity determined in this manner does not have the temperature dependence (T^2) expected from electron-limited phonon conduction.

The problem with the above analysis is that in strong fields the large lattice conductivity determined from Eq. (5) would result in a substantial decrease (~40%) in the measured Righi-Leduc coefficient [Eq. (4)]. As previously reported,[5] no such large decrease is observed and from the smaller decrease that was observed (5% at 9 T) a value of k_g can be extracted that has the correct temperature dependence but is somewhat smaller than the theoretical prediction (curve III, Fig. 6). The smaller magnitude of k_g could very well be due to dislocation and strain-field scattering which may be shown[25] to yield a thermal resistivity proportional to T^{-2}. As k_g has a very small effect on the Righi-Leduc coefficient, the error in determining k_g is large and we estimate it to be of the order of 50%; the sample to sample variation in k_g is also about 50%. The lattice conductivity determined from specimen KHF-5 is close to the average found for specimens KHF-5, 7, and 8, and was used to correct the thermal-magnetoresistivity data for specimens KHF-1 and 2, on which no Righi-Leduc data were taken. The lattice correction to the magnetoresistivities was done in the following manner. For specimens KHF-5, 7, and 8 we used the technique described in Ref. 5, i.e., we treated k_g as a free parameter and adjusted it until Eqs. (3) and (4), when solved simultaneously, yielded the observed Righi-Leduc data. We then extract $W^e_{xx}(H,T)T$ easily. For specimen KHF-1 and KHF-2 k_g and R_{RL} were chosen to be $3.3\times10^{-2}\ T^2$ W/m K and 1.76×10^{-2} m^3 K^2/W Ω C (from Ref. 5), respectively. Equations (3) and (4) then yield $W^e_{xx}(H,T)T$.

The electronic thermal magnetoresistance $W^e_{xx}(H,T)T$ was plotted versus field in a manner similar to Fig. 2. It is found that the various curves at the different temperatures are generally shifted up or down depending on the values of H and T, the change seldom exceeding 10% at 9 T. Similarly, when $\Delta W/H$ vs H is plotted the data are still very well represented by a polynomial to second power in H,

$$W^e_{xx}(H,T)T = W(0,T)T + A^eH + B^eH^2 . \quad (6)$$

The values of A^e and B^e are listed in Table II for all the specimens, and the behavior of the coefficients A^e and B^e is discussed in Sec. IV B.

There are two additional points worth pursuing. Archibald, Dunick, and Jericho[26] have measured the lattice conductivity of potassium directly, suppressing the electronic conductivity by alloying. Their specimens were heavily doped with rubidium. For their purest alloy specimen (Ref. 26, Fig. 4) they obtain a lattice thermal conductivity which appears to have the temperature dependence necessary to explain our thermal magnetoresistivity data (i.e., k_g is similar to Fig. 6, curve I) but with a magnitude two to three times too small. It is nevertheless several times larger than the magnitude we obtain[5] for k_g. Although their results are intriguing, we do not feel that they are applicable to the problems discussed in this paper. The atomic mass of rubidium is twice that of potassium, and this large mass difference results in strong Rayleigh scattering and thus strong phonon-impurity scattering. This scattering causes k_g to depart from the T^2 behavior because of electron-phonon scattering at the higher temperatures. Our specimens, however, are much purer than those of Archibald *et al.* (our least pure specimens have a RRR 50 times greater than their purest alloy) and such scattering should be minimal. The fact that the k_g obtained from the Righi-Leduc data is essentially independent of purity[5] can be interpreted to mean that the samples are sufficiently pure so that phonon-electron and phonon-boundary or dislocation scattering is dominant. However, decreasing the impurity content should increase the magnitude of k_g, beyond that measured by Archibald *et al.*, perhaps to a size sufficient to explain our magnetoresistance data. But a much larger k_g would have resulted in a much larger field dependence to R_{RL}. An additional important observation

is that the measurements of Archibald *et al.* were performed on encapsulated specimens. This method is now considered unreliable because of the large pressure dependence of the resistivity of potassium and the very real possibility of differential contraction between the specimen and the capsule. It must be stated, however, that the k_g inferred from R_{RL} by Tausch and Newrock and that measured by Archibald *et al.* are in disagreement.

The second point is that Eqs. (3) and (4) are sensitive to small changes in W^e_{xy}. For example, Eq. (4) may be expanded and solved for k_g,

$$k_g = \frac{1}{W^e_{xx}} \left(\frac{W^e_{xy} - W^M_{xy}}{W^M_{xy}} \right) . \tag{7}$$

Thus the value obtained for k_g depends on the value chosen for W^e_{xy}, the electronic contribution to the Righi-Leduc coefficient. One might start with the free-electron value, as predicted by the Lifshitz-Azbel-Kaganov (LAK) theory.[19] If however, the actual Righi-Leduc coefficient in potassium differs from the free-electron value, as is also true in the electrical case[27] where R_H is larger than the free-electron value $R_{H,fe}$ by about 5%, then large errors in k_g will result, eventually leading to large errors in $W^e_{xx}(H, T)$. To avoid this pitfall in part, Eq. (4) was fit to the Righi-Leduc data in the intermediate-field region (2–6 T) where the effects of lattice conduction are small. However, the possibility of large errors in k_g certainly exists and, coupled with an incorrect W^e_{xy}, could lead to large errors in $W^e_{xx}(H, T)$. To obtain a measure of these errors we have calculated $W^e_{xx}(H, T)T$ from typical $W^M_{xx}(H, T)T$ data, using our best estimate for k_g and W^e_{xx}, as well as values 50% too large or too small, and values of W^e_{xy} 10% above and below the free-electron value. From calculations of all possible permutations we are able to conclude that (a) at low temperatures ($T \approx 2$ K) the maximum error in $W^e_{xx}(H, T)T$ produced by a 50% change in k_g and a 10% change in W^e_{xy} is less than 0.4%. Similarly, the errors in the coefficient A^e and B^e are negligible; and (b) at the higher temperatures reached ($T \sim 8$ K), a ±50% change in k_g and a 10% change in W^e_{xy} produces at most a 6% shift in $W^e_{xx}(H, T)$, and a similar error in A^e and B^e. Since we estimate that our determination of k_g is correct to 50%, we feel the error in A^e and B^e attributable to this calculational problem is never more than 6%, and is generally much less. We note that the errors in A^e and B^e due to the error in determining the geometrical factors are larger.

We conclude therefore that the quadratic term in the measured thermal magnetoresistivity is not due solely to lattice conduction.

C. Measurement artifacts

There remains the possibility that the quadratic term is an experimental artifact. As far as we are able to determine there are three possible sources of error in our measurement that could lead to a quadratic term, or, at the very least, to an incorrect measurement of the thermal resistivity. These are a leakage conductance across the specimen, a thermometry error, and probe or aspect-ratio effects. We discuss these in order.

1. Leakage conductivity

Any field-independent thermal conductance which allows a thermal current to flow across the specimen will result in a quadratic term in the thermal magnetoresistance. This occurs in a manner similar to that of the lattice conductivity. The only possibilities in our experiments are the thermal conductance of the residual gas, the oxide layer on the specimen itself, and the thermometer wires. It is highly unlikely that any of these cause a significant effect. The vacuum in our cryostat is of the order of 10^{-8} Torr, and a simple calculation of the conductance of the residual gas (helium) indicates that it is much too small to cause the observed quadratic term. More importantly, in an earlier experiment the vacuum in the cryostat was varied between 10^{-5} and 10^{-8} Torr and no pressure-dependent effects were observed. The oxide layer is very nonuniform and quite thin; since it is an insulator it is quite difficult to imagine that it has any appreciable thermal conductance. Finally, there is 5 Ω of resistance wire separating the various thermometer leads from one another and from the heat stations. This is more than enough to eliminate any thermal leaks. Perhaps the most compelling argument against leakage conductivity may be found in the Righi-Leduc data.[5] If leakage conductances are to have a noticeable effect they must be at least of the same order of magnitude as the lattice conductance. Such leakage conductances would add to the lattice conductance, creating an effective lattice conductance, which would be the quantity determined above [Eqs. (3) and (4)]. However, the k_g we determine is *less* than the theoretical expectation, not greater. In addition the lattice conduction determined above has the quadratic temperature dependence typical of phonon-electron scattering. Thus it is unlikely such leakage is present, but should such conductances be present they would be too small for an observable effect on $W^e_{xx}(H, T)T$.

2. Thermometry

The resistances of the germanium thermometers used in this work increase with field roughly as

H^2; thus the possibility exists that the quadratic term might be due to calibration effects peculiar to our measurement techniques. To eliminate the possibility, the thermal magnetoresistance of a potassium specimen has been measured in a constant field using different heat currents. The size of the thermal gradient varies with the heat currents and moves the hot thermometer resistance farther from or nearer to its value during the calibration run. We found the thermal magnetoresistance to be independent of the magnitude of the heat current. In addition, with the same thermometry and calibration techniques, no quadratic term was observed in the longitudinal thermal magnetoresistance. We note that Fletcher,[18] using different techniques, has also observed the quadratic term.

3. Probe effects

In magnetoresistance measurements it is extremely important to consider the placement of the potential and current probes carefully. It is well known, for example, that a poor choice of aspect ratio will result in a magnetoresistance that is linear in the magnetic field.[28] In this case we must consider the effects on the thermal magnetoresistance measurements of the placement of the measuring thermometers, the injection and extraction of the thermal current, and the geometrical arrangement of the specimen.

In the case of the electrical magnetoresistivity the problem of the distortion of the equipotentials by the voltage and current probes has been investigated by several authors.[29] Among other possibilities Jensen and Smith[29] consider the case of point voltage contacts with electrical current injection and extraction via perfectly conducting contacts that completely cover the ends of the specimen. They demonstrate that for $\omega_c\tau = \infty$ most of the distortion occurs within a distance from the ends of the specimen equal to the specimen's width. If the voltage probes are away from the ends of the specimen by a distance equal to the specimen's width they show that the error in the electrical magnetoresistivity measurement is less than 0.1%, and decreases rapidly as the distance increases. If the current is injected and removed through sufficiently high impedances these effects are substantially reduced. The flow of thermal current is similar to the flow of electrical current. That is, if we inject and extract the heat current through perfect thermal conductors, and if the measuring thermometers are more than a specimen's width away from the current probes, the distortion of the isotherms by the current probes should result in an error in the thermal magnetoresistance of less than 0.1%, and the error will also be substantially reduced by using high-impedance heat-current injection and extraction. In our experiments the potassium specimen is clamped at the cold (platform) end, and a tweezer heater is fitted to the hot end.[1] The heater is thermally attached to the specimen by a thin film of DC-200 fluid which is glassy at low temperatures. We feel that this is certainly "high-impedance" current injection and extraction. In addition, in some of our measurements the measuring thermometers were closer to the ends of the specimen than the width, and in some cases several times as far away. Over the past several years a wide variety of specimens with various purities and aspect ratios have been measured; the consistency of the data is convincing evidence that there are no probe effects in the data.

In sum, we are reasonably certain that the quadratic term is not due to lattice conduction, to probe effects, or to any other spurious effect, but that it is an intrinsic part of the thermal magnetoresistance of potassium. However, as there are no probeless methods for measuring thermal resistivities, we cannot completely exclude the possibility of some form of systematic exotic probe effect.

There are several important criticisms of these arguments that should be mentioned. It is entirely possible that the dip in TR_{RL} with field which we previously reported[5] is not due to the lattice conductivity but is due to an intrinsic field dependence of R_{RL}. This possibility cannot be discounted, particularly since field-dependent Hall coefficients have been observed and predicted by theory (see below). Second, in obtaining the k_g used to correct our measured values of the thermal magnetoresistivity, we assumed that W^e_{xy} was nearly equal to the semiclassical prediction. We can also reinvert the resistivity tensor and obtain

$$k_{xy} = (-1/W^M_{xy})/[1 + (W^M_{xx}/W^M_{xy})^2].$$

If our values for W^M_{xx} and W^M_{xy} are inserted in this equation, k_{xy} is *not* field independent as predicted, and our original assumption is violated. There are a number of possible explanations.

(i) Our R_{RL} data are incorrect. This is very unlikely. Aside from the various precautions mentioned above we have measured R_{RL} for aluminum and have reproduced the expected results.

(ii) The inversion of the resistivity tensor to obtain conductivity tensor elements is incorrect. This is the most likely explanation. Without an adequate knowledge of the causes of the magnetoresistance anomalies and the way in which the conductivity tensor elements are affected such an inversion is improper and can lead to error. The most obvious case is an extrinsic mechanism

which contributes to the effective resistance, such as sample inhomogeneity or end effects. Clearly, if such mechanisms are present conductivity tensor elements cannot be calculated correctly from the resistances. Thus without knowing the ultimate source of the magnetoresistance anomalies such inversions must be viewed with some suspicion. Note however, that the fact that W_{ij} cannot be properly inverted can be interpreted in a manner which lends credence to claims that the magnetoresistance anomaly is extrinsic.

(iii) The results of Archibald *et al.*[26] can be interpreted as supporting Fletcher's argument[18]; that is, the entire quadratic term may be due to lattice conduction. This of course leaves unexplained the behavior of the Righi-Leduc coefficient.

We also observe that the lattice conductivity does not produce a quadratic term in the longitudinal magnetoresistivity and the absence of such a term in the data is also interpretable as supporting the extrinsic hypothesis.

V. DISCUSSION

In this section we discuss the thermal magnetoresistance of potassium, taking two somewhat different approaches. In the first the linear term in Eq. (2) is investigated and correlations between that term and the linear electrical magnetoresistivity are examined. Here the quadratic term with its strong temperature dependence is treated as an additional anomaly. Much of this discussion is similar to that of Ref. 1, except that now lattice conduction effects are included. The second approach is an examination of the thermal magnetoresistance in its entirety. Several important effects are noted.

A. Linear term

In Fig. 7 the coefficients of the linear term [Eq. (2)], A and A^e corrected and uncorrected for K_g, are plotted versus the cube of the temperature for specimen KHF-8. The data are represented very well by $A^e = A_0^e + A_1^e T^3$ to temperatures of nearly 8 K. The values of A_0^e and A_1^e for all the specimens are listed in Table II. In general we observe that by correcting the data for the lattice conductivity we may extend the apparent T^3 behavior to higher temperatures. The deviation of A^e from T^3 behavior at high temperatures is not due to leaving the high-field limit, as may be observed from the values of H_1 listed in Table II.

There are several interesting points concerning the temperature and purity dependence of A^e. As may be observed from Table II the temperature-independent portion of the linear term A_0^e is purity dependent. Thus this term appears to be directly

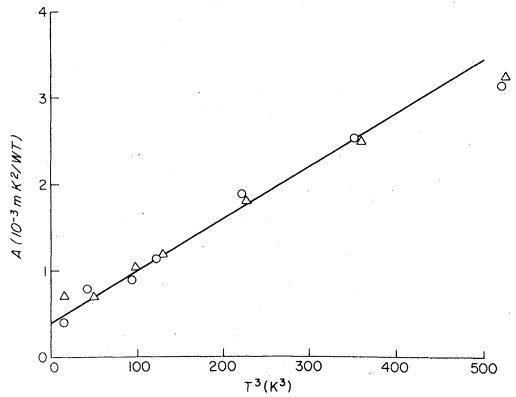

FIG. 7. Coefficient of the linear term [Eqs. (2) and (6)] vs the cube of the temperature for specimen KHF-8; corrected (Δ) and uncorrected (○) for the lattice conductivity.

related to the impurity content and defect structure of the specimen. Following our previous procedure[1] we find that the linear term in the thermal magnetoresistivity can be expressed in terms of a thermal Kohler slope S_T

$$\left.\frac{\Delta W(H,T)}{W(0,T)T}\right|_{\text{linear}} = \frac{A^e H}{W(0,T)T} = A^e \frac{L_0}{R_H}\omega_c\tau = S_T(\omega_c\tau). \tag{8}$$

If we assume that the semiclassical prediction for the off-diagonal elements of the resistivity tensors is valid (it is to within a few percent), the zero-temperature Kohler slope may be written as $S_T^0 = A_0^e/R_{RL}T$. We also assume that the Wiedemann-Franz law is obeyed for the off-diagonal elements, at least to within 5%–10% (Refs. 1 and 27). Since we are only interested in rough comparisons between the thermal and electrical cases such errors are unimportant. Using[5] $TR_{RL} = 1.76 \times 10^{-2}$ $m^2 K^3/W\Omega C$ we may now determine S_T^0, and this is listed in Table II, along with the electrical Kohler slope for specimens of similar purity. The thermal slope is usually three to four times larger than the electrical slope. In Ref. 1 it was pointed out that strain and dislocations in the specimens may be responsible for this difference. Precautions were taken to limit the strain induced in mounting and precooling the specimens. However, our method of sample preparation (pressed plates) may be considered a "high-strain" technique.[30] As noted in Ref. 1 the linear electrical magnetoresistance of one of our pressed specimens was quite large, indicating the presence of strain. That specimen was frozen in place by oil for helicon measurements; thus it is possible that the electrical measurement was on a strained specimen, whereas the thermal measurement was

not. Also, as discussed above, the lattice conductivity varies as T^2, but has a magnitude less than the theoretical prediction; strains and dislocations can produce such an effect.[25] On the other hand the data from the single-crystal specimens used in Ref. 1, fabricated using a "low-strain" technique, are quite consistent with the data from the polycrystals.

The nature of the purity dependence of the two Kohler slopes is similar. In Figs. 8(a) and 8(b) S_T and S_E are plotted vs RRR. Although the number of thermal data points is insufficient for any firm conclusion, the purity-dependent portion of the linear thermal magnetoresistance appears to depend on purity (as determined by the residual resistivity) in much the same manner as the linear electrical magnetoresistance. This is illustrated by the large peak in the Kohler slope for a RRR of 1700. Figure 8(c) is a plot of the purity dependence of the phonon term in the zero-field thermal resistivity [B_0, Eq. (1)]. (The data are from Refs. 1, 3, and this work; see Ref. 3 for the details.) We offer the following speculation on the origin of the large peak in S_E and S_T^0. B_0 is essentially constant for very pure or very impure specimens (as determined by their RRR), and rapidly changes in the intermediate-purity regime. This variation with purity of the magnitude of the temperature-dependent portion of the zero-field thermal resistivity is a form of deviation from Matthiessen's rule.[3,31,32] The response of the electron distribution function to the applied electric field is changing from being dominated by impurity (or strain field) scattering (at low RRR) to being dominated by phonon scattering (at high RRR). From a comparison of Figs. 8(a) through 8(c) we see that the change in B_0 occurs over the same purity range in which both S_T and S_E rise, reach their peaks, and fall. Therefore, the purity dependence of the thermal and electrical linear magnetoresistances might simply be manifestations of deviations from Matthiessen's rule and not directly related to the cause of the magnetoresistance anomalies.

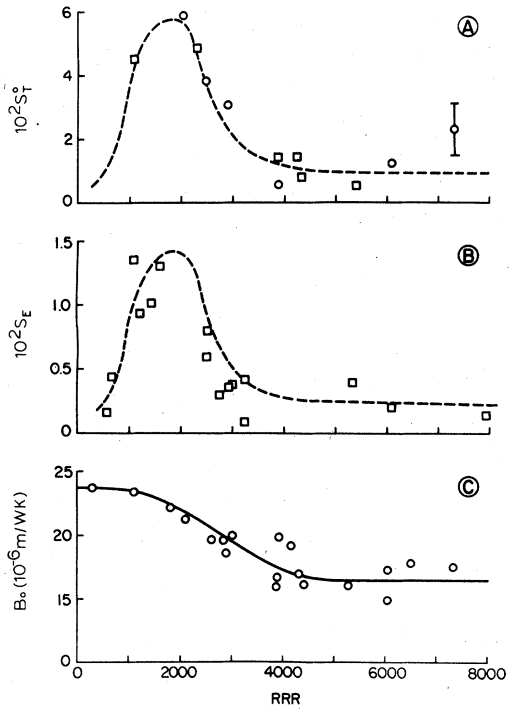

FIG. 8. (a) Kohler slope of the transverse electrical magnetoresistance as a function of purity as measured by the RRR (from Refs. 17 and 32). (b) Zero-temperature Kohler slope of the linear term in the transverse thermal magnetoresistivity as a function of purity (from Ref. 1 and this work). (c) The coefficient B_0 [Eq. (1)] as a function of purity (from Ref. 1, 3, and this work). In all three figures the lines are drawn to indicate the trend of the data and have no further meaning.

Since the temperature-dependent term in the zero-field thermal resistivity shows substantial deviation from Matthiessen's rule. It is natural to look for such deviations in the temperature-dependent term of the linear magnetoresistivity A_1^e. As seen in Table II, no such effects are observable within the experimental uncertainty.

Reference 1 describes a relationship between the temperature dependence of the linear thermal magnetoresistivity, the zero-field thermal resistivity, and the analogous terms in the electrical resistivity. The temperature-dependent portion of both the zero-field thermal resistivity ($W_T T = W(T,0)T - [W(T,0)T]_{T=0}$) and the linear term in the thermal magnetoresistivity have the cubic temperature dependence typical of phonon scattering, and the temperature-dependent portions of the zero-field electrical resistivity and the linear electrical magnetoresistivity also have the same temperature dependence.[1] This is also apparent in the data presented here, and we therefore reiterate the conclusion of Ref. 1: "It appears that the temperature dependent portions of the linear magnetoresistances are related to the strength of the electron-phonon interaction (as measured by the zero-field resistivities) even though the relative contributions of large and small angle scattering are much different in the two cases."

B. Quadratic term

In very strong fields ($\omega_c \tau \gg 1$) the quadratic term dominates the transverse thermal magnetoresistivity. There is no analogous term in the transverse electrical magnetoresistivity. Figure 9 shows a plot of $\ln B$, the coefficient of the quad-

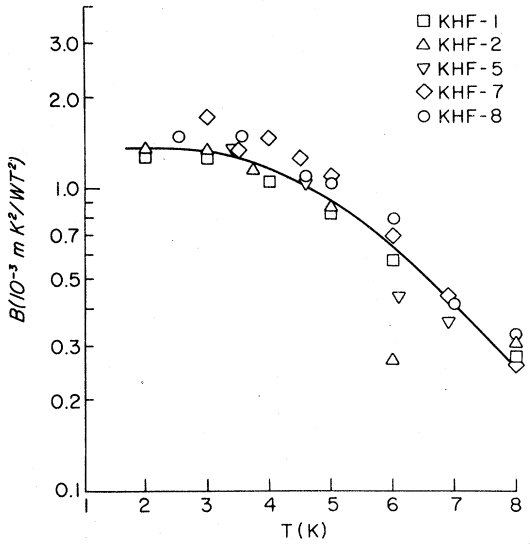

FIG. 9. B^e [from Eq. (6)] vs the temperature. B^e is seen to be a monotonically decreasing function of temperature with no simple temperature dependence.

ratic term vs T for all the specimens. There is no simple power law dependence. B is seen to be a monotonically decreasing function of T and, from Table II, may be seen to be essentially independent of purity (within the experimental error) although KHF-7, which has a Kohler slope at the peak of Fig. 8(b), appears to have the largest B.

From our discussion it is clear that the quadratic term cannot be ascribed solely to lattice conduction, and that lattice conduction can only account for a small portion of it. We are unable to offer any explanation of the origin of this term, but assume it to be related to whatever is causing the linear anomalies. It should be noted, however, that this term causes drastic deviations from the Wiedemann-Franz law. Since the high-field transverse thermal magnetoresistivity is quadratic in the field, whereas the transverse electrical magnetoresistivity is linear in the field, the Lorenz ratio decreases with increasing field,

$$\frac{\rho(H,T)}{W(H,T)T} = \frac{\rho(0,T)+\Sigma H}{W(0,T)T + A^e H + B^e H^2} \xrightarrow[H\to\infty]{} \frac{\Sigma}{B^e}\frac{1}{H}. \quad (9)$$

Σ is very nearly temperature independent,[33] whereas B^e increases rapidly as the temperature decreases (Fig. 9) and the deviation from L_0 is largest at low temperatures. In the low-temperature regime elastic impurity scattering is expected to be the dominant scattering mechanism, and this deviation is difficult to understand.

C. Kohler's rule

To this point the linear and quadratic terms in the thermal magnetoresistivity have been discussed separately. In this section they are treated together, and a method of analyzing the data is presented that appears to eliminate much of the complicated temperature dependence. This may shed some light on the ultimate source of the anomalous magnetoresistivities.

Using the methods outlined in Ref. 5, we may obtain values for the thermal scattering time τ_{th}; using these values we then investigate changes in the transverse thermal magnetoresistivity as a function of temperature at constant values of $\omega_c\tau_{th}$. We proceed as follows: from the known relation between τ_{th} and T the magnetic field necessary to yield a given value of $\omega_c\tau_{th}$ (at a given temperature) is determined. We obtain the transverse thermal magnetoresistance at that field and temperature from the measured values of $W(H,T)$ or from Eq. (6). Repeating this at a variety of fields and temperatures for each value of $\omega_c\tau_{th}$ we plot the results in Fig. 10 for specimen KHF-5 (similar data for KHF-1 have been published elsewhere[16]). This figure displays the thermal magnetoresistivity times temperature at constant $\omega_c\tau_{th}$, $W(\omega_c\tau_{th}=c)T$, versus the cube of the temperature. Also included in the figure is the zero-field thermal resistance data for the specimen, $W(0,T)T$.

As may be observed in Fig. 10, for specimen KHF-8, as well as for all the specimens, over the entire purity range investigated $W(\omega_c\tau_{th}=c)T$ is a cubic function of the temperature over a range of values of $\omega_c\tau_{th}$ from zero to at least 90. The complicated temperature dependence of the thermal magnetoresistivity (as shown in Figs. 1 and 2) has been eliminated. There is none of the crossing behavior present, and little or no deviation

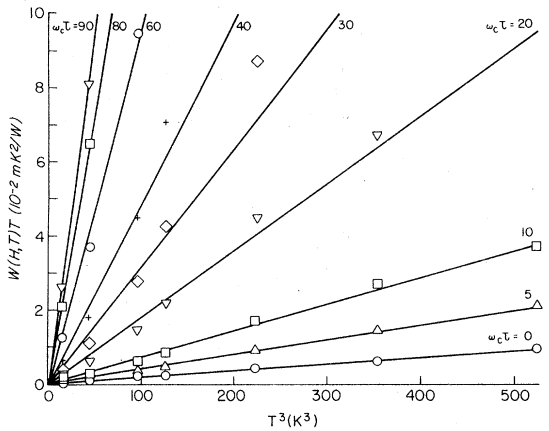

FIG. 10. Thermal magnetoresistivity at constant $\omega_c\tau_{th}$ times temperature as a function of the cube of the temperature [$W(\omega_c\tau_{th}=c)T$ vs T^3] for various values of $\omega_c\tau_{th}$, specimen KHF-8.

from T,[3] except possibly at the lower temperatures. At low temperatures the percentage error in the value τ_{th} is considerably larger than at the higher temperatures, and because of the large quadratic field dependence of $W(H,T)T$ the calculation of $W(\omega_c\tau = c)T$ at low temperatures is quite sensitive to small changes in $\omega_c\tau_{th}$.

This is a very striking and surprising result. Although the coefficient of the linear term is a cubic function of the temperature the coefficient of the dominant quadratic term has no simple temperature dependence, and $W(H,T)T$ has no simple temperature dependence.

In Fig. 11 the slopes (Γ) of the curves in Fig. 10 as well as the slopes of similar curves for the other specimens are plotted versus $\omega_c\tau_{th}$. Except for specimen KHF-7 the slopes have nearly the same dependence on $\omega_c\tau_{th}$. This is consistent with the results discussed above for the purity dependence of the slope of the linear terms. Sample KHF-7 has a RRR of 2090, nearly at the top of the peak in Fig. 8(a) and in the middle of the rapid variation of B_0 in Fig. 8(c) the other specimens have RRR's on the flat parts of those curves. We observe that scaling the KHF-7 data by the proper ratio of B_0's produces, within experimental error, a universal curve. (The tips of the arrows in Fig. 10 show the location of the scaled points.) This is further evidence of the possible effects of deviations from Matthiessen's rule.

The fact that $W(\omega_c\tau = c)T$ is a cubic function of the temperature for all $\omega_c\tau_{th}$ including zero indicates that Kohler's rule holds. A thermal Kohler's rule[34] may be defined as

$$\Delta W(H,T)/W(0,T) = F(\omega_c\tau_{th}), \tag{10}$$

or

$$W(H,T) = W(0,T)[1 + F(\omega_c\tau_{th})]. \tag{11}$$

Since $W(0,T)T$ is a cubic function of the temperature over a wide range of temperatures,[1] the fact that $W(\omega_c\tau_{th} = \text{const})T \propto T^3$ means that Kohler's rule is valid, at least for the range of fields and temperatures investigated here. The fact that Kohler's rule holds is somewhat surprising: at the lower temperatures of our measurements the electrons are primarily scattered by impurities, while at higher temperatures they are predominately scattered by phonons.

This is an extremely important result and has significant consequences. The domain of validity of Kohler's rule has not been delineated other than in the simple relaxation-time approximation. It can however, be shown that if anisotropy is significant (e.g., with open orbits or magnetic breakdown), there will be large deviations from

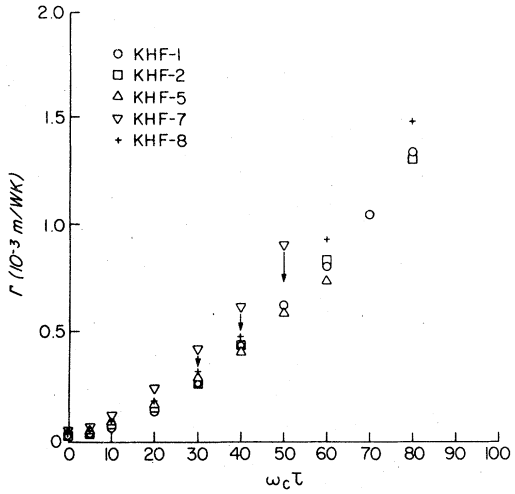

FIG. 11. Slope (Γ) of the curves in Fig. 10 along with the slopes of similar curves for the other specimens. The tips of the arrows (for specimen KHF-7) indicate the values obtained when the slopes are scaled by the ratio of the coefficients B_0 (see text) to correct for possible deviations from Matthiessen's rule.

Kohler's rule. It will be violated if the field dependence of the magnetoresistance is not due solely to the curvature of the electron orbits about the field direction.[35] This has important consequences in the creation of a correct theory of the magnetoresistance anomalies. In particular, any theory of the anomalous magnetoresistances of the simple metals which is based on the introduction of open orbits or magnetic breakdown cannot be correct.

Of all the experimental observations discussed in this paper and elsewhere, several stand out as being crucial to understanding the magnetoresistance problem. They are (i) the nonsaturation of either of the longitudinal magnetoresistances, (ii) the large deviation from the Wiedemann-Franz law at low temperatures, (iii) the incorrect magnitude of the Hall coefficients, (iv) the Kohler's rule scaling of the thermal magnetoresistance. Using these observations we are able to eliminate many of the proposed mechanisms and this will be discussed further elsewhere.[36]

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*Present address: Libbey-Owens-Ford, Inc., Toledo, Ohio.
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