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Righi-Leduc- and Hall-Coefficient Measurements on Na and K[†]

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Experimental results on the Righi-Leduc and Hall coefficients of K and Na over the range 2–100 K are reported. The Righi-Leduc coefficient does not behave as expected but shows a large peak at low temperatures; the origin of this peak is not clear but it may be due to phonon drag. The measurements have been made over a wide field range (0.05–0.95 T) and it is found that the limiting low-temperature behavior is sensitive to field. At low temperatures and high fields, the Wiedemann-Franz law is found to hold to within 1%, which suggests that the thermal conductivity of the lattice must be small in comparison with that of the electrons; in addition, the Righi-Leduc and Hall coefficients exhibit the expected magnitude appropriate to a closed electronlike Fermi surface.

I. INTRODUCTION

We have recently reported[1] our experimental results on the Hall coefficient R and its thermal analog the Righi-Leduc coefficient A of the pure noble metals over the temperature range 4–120 K. (Reference 1 is referred to as I in the following.) If the electrons are scattered elastically and if the lattice conductivity is negligible compared to the electronic thermal conductivity, then R and A should be related by the Wiedmann-Franz law,

$$R = A L_0 T, \tag{1}$$

where L_0 is the Lorenz number and T the absolute temperature. In the case of the noble metals the results were relatively complicated, since both R and $A L_0 T$ were strongly temperature dependent and the distorted Fermi surfaces of these metals permits only approximate theoretical treatments. The temperature dependence of R has been semiquantitatively explained on the basis of a two-band model, but we were not able to extend the treatment satisfactorily to $A L_0 T$. To be specific, the model suggests that the maximum (absolute) magnitude of $A L_0 T$ should not exceed R, and this was not found to be the case experimentally. Considerable simplification in the analysis can be made if the Fermi surface is spherical; thus we undertook a series of measurements on the alkali metals and in this paper we report our results on Na and K. The data of Alderson and Farrell[2] shows that R depends only weakly on temperature for these metals and they are thus ideal for our purposes.

In Sec. II we present our data and outline the experimental techniques. In Sec. III we discuss the results in the light of available theories.

II. EXPERIMENTAL TECHNIQUES AND RESULTS

Dugdale and Gugan[3] have shown that measurements made on alkalis that have been cast into tubes are unreliable indicators of the behavior of unconstrained samples. With this in mind, we decided that our measurements should be made on essentially self-supporting specimens as far as was practical. The alkalis, however, are so soft that, in the sheet form that we require, they are not capable of completely supporting themselves and the heaters, thermocouples, etc., that are necessary for these experiments. To overcome this difficulty, the mounting frame of Fig. 1 was constructed. The samples were clamped to copper blocks at the ends of the frame as indicated; this was done in such a way that a copper foil was

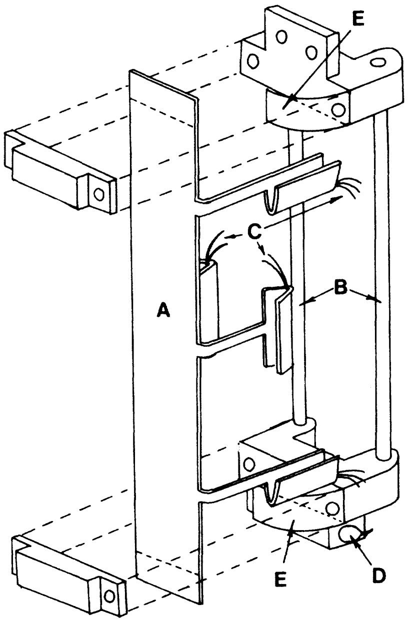

FIG. 1. Method of mounting the Na and K samples. A: sample; B: threaded nylon rods; C: thermocouples and other leads crimped in the limbs of the specimen; D: heater; E: copper foil sandwiched between a fold in the end of the specimen to improve thermal and electrical contact. The heat conduction through the nylon rods is totally negligible for the specimens used in this work.

sandwiched within a fold at each end of the specimen to ensure good electrical and thermal connection. When the Hall coefficient was to be determined, the sample was supported against the Lorentz force by two Perspex sheets that could be attached to the frame with ample clearance to prevent squeezing the specimen.

The alkalis were obtained from various sources indicated in Table I, and were formed into flat sheets by rolling with a stainless-steel roller and block. The thickness was controlled by shims along the edges of the block, and a razor blade and template were used to cut the shapes required. Before mounting, the samples were always annealed by storing them under dried oil for at least one day. After this time the crystallites of the sample were quite noticeable and were typically 1 mm in size. Thermocouples and electrical leads were clamped between folds of the specimen limbs shown in Fig. 1.

Because of the reactivity of the alkalis, all sample handling was carried out in a glove box with an argon atmosphere. Usually the samples were covered with a thin film of oil, which, together with the inert-gas atmosphere, prevented any significant corrosion of the surfaces. The oil used for this and for storing the samples was previously dried by stirring molten sodium or potassium through the oil for about 12 h. The atmosphere in the glove box could only be maintained in a relatively clean condition by continually exposing freshly cut Na or K surfaces; this was the case even though the box was leak tight and an overpressure of argon always maintained. The cryostat, and anything else required in the glove box, was inserted through an evacuable port to prevent the entry of air.

TABLE I.

Sample	Source[a]	$\sigma_{4.2}/\sigma_{295}$	$\omega_c\tau$ at 4 K and $B=1$ T using $\omega_c\tau = R\sigma B$	Parameter(s) measured
Na3	Baker	2700	13.4	A_m, R
Na4	Mackay	1500[b]	7.5	A_m, R
Na5	Baker	6700[c]	33	A_m, R
Na8	Baker	1620	8.0	A_m, R
K1	Baker	720	4.4	A_m
K2	Mackay	2900	18	R
K4	Baker	700[b]	4	A_m
K5	M. S. A.	>2000[b]	>12	A_m
K6	M. S. A.	7900[d]	49[e]	R
K8	Baker	640	4.0	R
K9	Baker	1005	6.2	R, A_m[f]

[a] Baker refers to J. T. Baker Chemical Co., Phillipsburg, N. J. Mackay refers to A. D. Mackay and Co., N. Y. M.S.A. refers to Mine Safety Appliances Ltd., Callery, Pa.

[b] Estimated from thermal-conductivity data.

[c] This is the value measured on the first cooldown. On cycling this specimen, the value decreased as explained in the text.

[d] This is $\sigma_{2.0}/\sigma_{295}$.

[e] $\omega_c\tau$ at 2.0 K.

[f] 2–20 K only as a function of magnetic field.

When finally assembled it was required that the specimen should be inside a vacuum-tight copper can which in turn was inside a second vacuum-tight brass can. Because of the low melting points of the alkalis, the standard Woods metal seal for the inner can could not be used, and a seal described by Mota[4] was utilized instead. In this design a mechanical joint is made by means of a screw thread and the vacuum seal is provided by a soap-glycerine solution smeared over the threads before assembly. We found this to be absolutely reliable during cooling and warming, provided that thermal shocks were not too large. After sealing the inner can in this manner (while inside the glove box), it was pumped to less than 5×10^{-2} torr and removed to enable the outer can to be sealed with Woods metal in the usual way. The apparatus was then cooled over 30 min to 77 K and evacuated to better than 10^{-6} torr; during all measurements this vacuum was maintained in the inner can.

The estimation of the thickness of each sample is the major source of uncertainty in the absolute value of the Hall and Righi-Leduc coefficients. Thicknesses were typically 1 mm and were determined with a micrometer; the samples were so soft that errors up to 5% are possible, though the reproducibility and absolute values of the data suggests that an accuracy of better than 3% was actually obtained. Since an important part of the present investigation is a comparison of R and A, it is preferable to make both sets of measurements on the same samples, to eliminate thickness uncertainties. This required the transfer of the samples between two cryostats, and was found to be difficult in the case of K. In only one case were both sets of data determined on the same K sample (K9) but there was no problem in the case of the relatively harder Na samples. The only other significant source of uncertainty in our measurements is the calibration of the Au + 0.07 at.% Fe versus Chromel thermocouples. We have recently recalibrated these thermocouples in magnetic fields of up to 4 T. The new data for the thermopower are within 1% of our previous values over the range 2–120 K, except in the interval 4–9 K, where there is a maximum difference of 4%; the values of the emf are such that the indicated temperatures differed by less than 0.1 K above 20 K and less than 1% below 20 K. These results justify the errors we have previously placed on the thermocouples and the new calibration enables us to place the following limits on the thermopower and emf. The thermopower should by accurate to 1% and the temperature (corresponding to the emf) to 0.15 K between 4 and 120 K. Below 4 K the uncertainty in

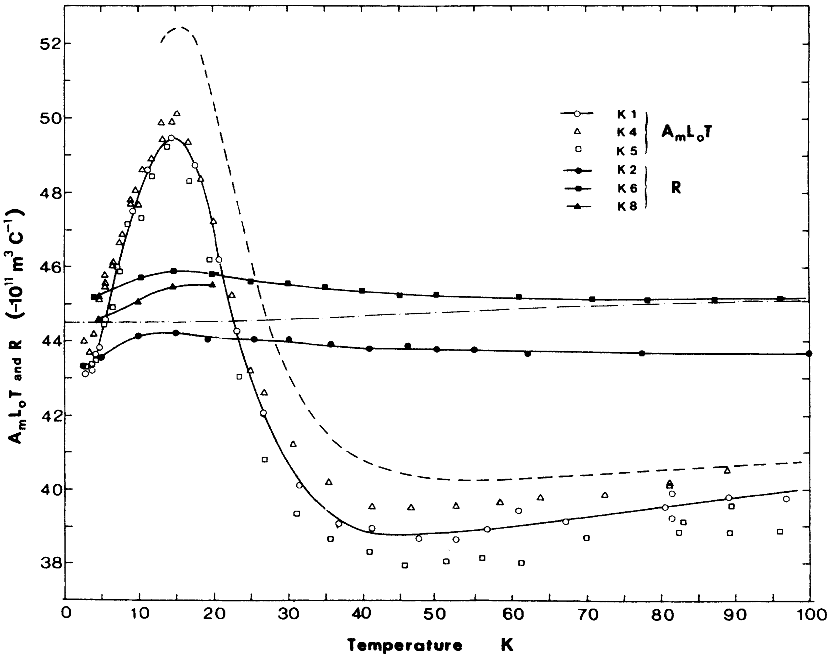

FIG. 2. Results on A_mL_0T and R for all the K samples except K9. The chain line (–•–•–•) indicates the theoretical value of $(ne)^{-1}$, which is slightly temperature dependent because of thermal expansion. The dashed curve (---) shows the estimated value of AL_0T after correcting A_mL_0T for the lattice conductivity as explained in the text.

the thermopower rapidly increases and could be as high as 5% at 2 K, though the indicated temperatures become more accurate. A magnetic field of 4 T changes the thermopower by less than 1% above 10 K, but by 4 K the thermopower has increased by 6%. We have restricted the field to less than 1 T in the present series of measurements and find changes in the thermopower of less than 1.5% at temperatures above 4 K. Below 4 K the effect of the field is relatively uncertain, and we have assumed the change in the thermopower remains 1.5% at a field of 1 T. Magnetic fields were determined by a Hall probe to better than 0.5%

The various samples we have used are listed with some pertinent information in Table I. We distinguish between the Righi-Leduc coefficient that we measure, A_m, and the coefficient that one would determine in the absence of the lattice conductivity λ_g, which we label A (see the discussion Sec. III A). The latter is the coefficient that we would prefer to evaluate, if it were possible, in order to compare with the theoretical results, e.g., Eq. (1). Both the A_m and R data were measured under adiabatic conditions (see I) and all the results have been corrected for the effects of thermal expansion.[5] Measurements were taken at two field values, 0.525 and 0.945 T. There was a noticeable difference in both R and A_m at the lower temperatures taken at the two fields, but Figs. 2–4 show only the measurements made at the higher fields; the behavior as a function of field will be returned to later.

All the data on K (except K9) are shown in Fig. 2. It is probable that the spread of the data mainly reflects uncertainties in the thickness of the samples, though it is noticeable that there tends to be more spread in the results for A_m at higher temperatures than at lower temperatures (this seems to be the case for both Na and K). The data for the Na specimens (except Na8) is shown in Figs. 3 and 4.

Dugdale and Gugan[6] have investigated the effects of the martensitic phase transformation of Na on the electrical resistivity. From their results they were able to show that the transformation shows considerable hysteresis. Thus on cooling a sample of Na, say from room temperature, there is no significant effect until about 30 K, when the sample begins to transform from the bcc to hcp phase. A fraction of possibly 20% may have converted by 15 K, below which no further change is evident. Warming the specimen leaves the fraction of the hcp phase essentially unchanged until about 45 K, when the Na begins to revert wholly to the bcc phase again, and the transformation is complete by about 70 K. Over the range 15–70 K there can thus be appreciable differences in the amount of hcp phase present, depending upon the previous history of the sample.

We have cycled our specimens in temperature in order to examine the effect of the hcp phase on the results. Figures 3 and 4 show that there is a much larger effect on A_m than on R; indeed we do not detect any effect at all on R within the experimental uncertainty. If one takes the specimen through a complete cycle without warming above 77 K, then

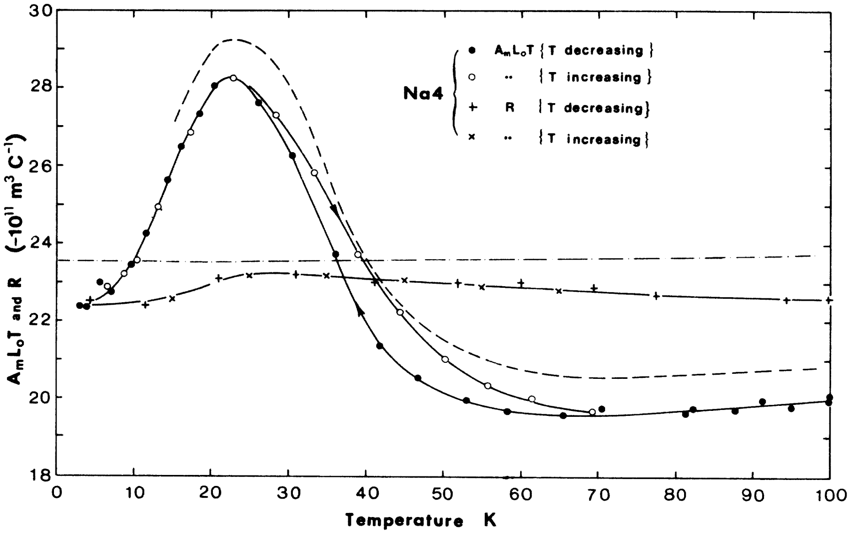

FIG. 3. Results on A_mL_0T and R for Na4. The chain line and dashed line are explained in the caption of Fig. 2.

it is found that on succeeding cycles less of the sample transforms to the hcp phase.[6] We have taken Na5 three times around such a cycle, and the results obtained on A for the first and third warming from 4 to 77 K are shown on Fig. 4. These results are clearly consistent with there being less hcp on the third cycle as compared to the first. It is interesting that there seems to be little effect at temperatures below the peak in A_m. Contrary to the experience of Dugdale and Gugan,[6] we saw large increases in the residual resistance of Na5 after cycling. Thus on the first cooldown the residual resistance ratio ($\sigma_{4.2}/\sigma_{295}$) was 6700; on the second, 4900; on the third, 4600; and on the fourth, 4400. Data for the other samples are not available, since they were cycled only once.

We have determined the thermal conductivity of our samples over most of the temperature range, though the high-thermal conductance of the samples rendered the results imprecise below 25 K for Na and below 15 K for K. In the case of Na the values we obtain for all the samples lie within 2% of those found by Cook *et al.*[7] over the range of overlap 37–100 K and exhibit the minimum in the region of 65 K. We find that K also shows a minimum at roughly 40 K, where we determine the thermal conductivity as 113 ± 3 W m^{-1}K^{-1}.

III. DISCUSSION

A. Low-Temperature Limiting Values

It has been shown that, providing the lattice thermal conductivity λ_g is negligible in comparison with the electronic thermal conductivity λ_e, and if the electronic scattering is elastic, then R and A are related by Eq. (1), whatever the magnitude of the magnetic field.[8] The discussion in I suggests

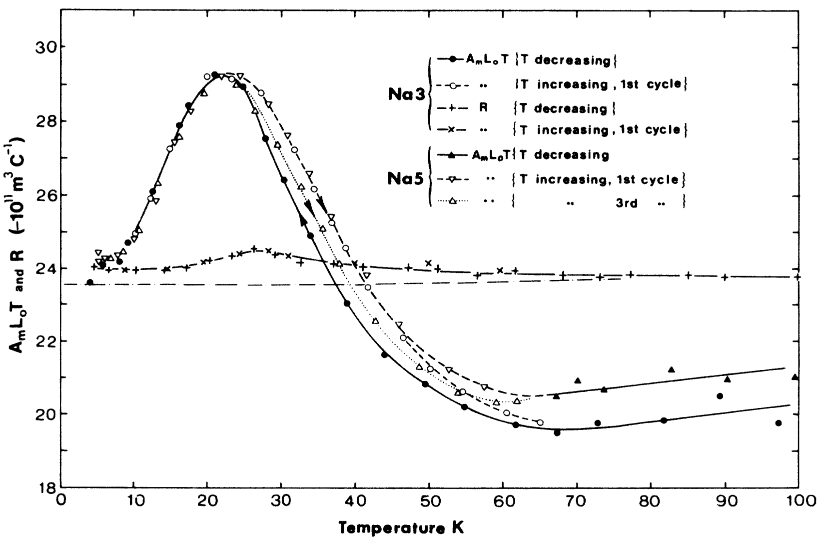

FIG. 4. Results on A_mL_0T and R for Na3 and Na5. The chain line is explained in the caption of Fig. 2.

that Eq. (1) will also hold, at least in low fields, even if the scattering is not elastic, but has the same anisotropy for the relaxation times associated with the thermal and electrical conductivities. Tsuji[9] has shown that if λ_g cannot be ignored, then the measured value of the Righi-Leduc coefficient A_m is related to the value A that would be measured in the absence of λ_g by the equation

$$A_m = A(\lambda_e/\lambda)^2, \tag{2}$$

where

$$\lambda = \lambda_e + \lambda_g .$$

The origins of this term are (also see the Appendix): (i) the electronic part of the thermal-current density is less than that measured by a factor (λ_e/λ) and (ii) the measured transverse-temperature gradient is smaller by the same factor, due to the thermal shorting effect of λ_g. In view of the origin of Eq. (2), it would be surprising if it depended on magnetic field, providing λ_e is measured in the magnetic field. We have tested Eq. (2) in the case of Ag alloys[10] and find that it holds when the electronic scattering is elastic, and we have every reason to believe that it will hold in all cases. Assuming the validity of Eq. (1) and if λ_g is not negligible, then we expect to find $R > A_m L_0 T$. At the lowest temperatures we should be in the elastic-scattering regime and it will be seen from Figs. 3 and 4 (and Fig. 6 for Na8) that our results at $B = 0.945$ T for Na satisfy[11] Eq. (1) to an accuracy of about 1%; it will be remembered that both R and $A_m L_0 T$ have been determined on the same samples in the case of Na, so that the comparison depends only on the thermocouple calibration and random errors. The comparison suggests $\lambda_g \lesssim 0.01\lambda_e$ at 5 K for Na. In the case of K, most of our data on R and $A_m L_0 T$ were not obtained on the same specimens. However, for K9 it was possible to determine both quantities, and the results for this sample (measured over the range 2–20 K) are shown in Fig. 5. In the high-field case[11] it will be seen that R and $A_m L_0 T$ are within 1% at the lowest temperatures. All the other K specimens in Fig. 2 show equality within the uncertainty due to errors in the thickness of the samples. Thus it again appears that $\lambda_g \lesssim 0.01\lambda_e$ in the region of 4 K for K. There has been speculation that λ_g might be a much larger fraction of the total thermal conductivity in this temperature range, particularly in the case of K. Archibald *et al.*[12] suggested that the transverse phonons should experience very little resistance, provided they were not strongly coupled to the longitudinal modes, and estimated $\lambda_g \sim \lambda_e$ at 2 K for large samples (i.e., not limited by boundary scattering). The results of Stauder and Mielczarek[13] were interpreted as evidence for λ_g being perhaps 25–50% of the total conductivity, but unfortunately the size of the samples is not given. The present data are inconsistent with λ_g being more than 1 to 2% of λ_e, though it should be remembered that we are working with polycrystalline samples with a grain size of roughly 1 mm.

Providing $\omega_c \tau \gg 1$ (ω_c is the cyclotron frequency and τ the electronic relaxation time), the Hall coefficient of a metal with a closed Fermi surface containing only electrons is given by

$$R = (ne)^{-1}, \tag{3}$$

where n is the number of electrons per unit volume and e the (negative) electronic chargs.[14] All our specimens are in the high-field regime at 4 K and 0.945 T (Table I lists values of $\omega_c \tau$ using the expression $\omega_c \tau = R\sigma_0 B$, where σ_0 is the electrical conductivity at 4.2 K), and we have plotted the values calculated from Eq. (3) on Figs. 2–4 and on Figs. 6 and 7. For Na, $(ne)^{-1} = -2.354 \times 10^{-10}$ m^{3}C^{-1}, and

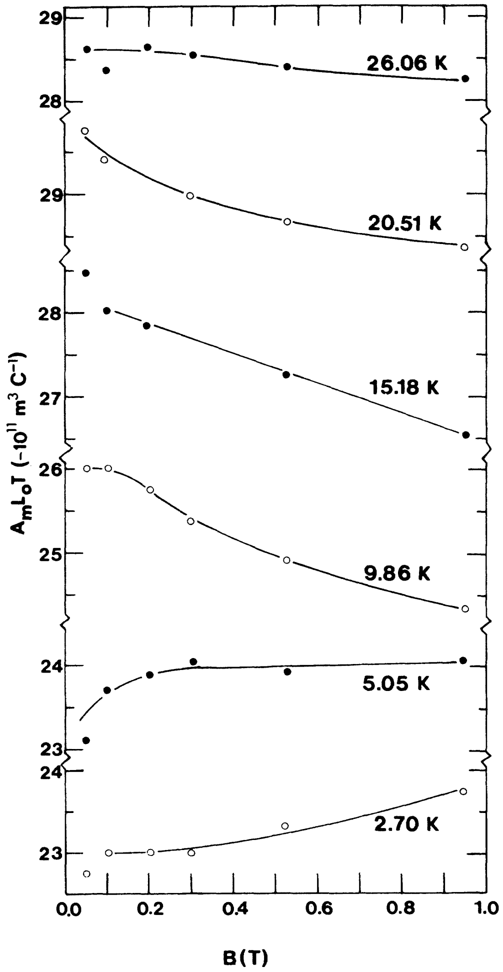

FIG. 5. Some of the data showing the effect of varying the magnetic field on $A_m L_0 T$ for Na8. The data for R are similar as are the data for both R and $A_m L_0 T$ on K9.

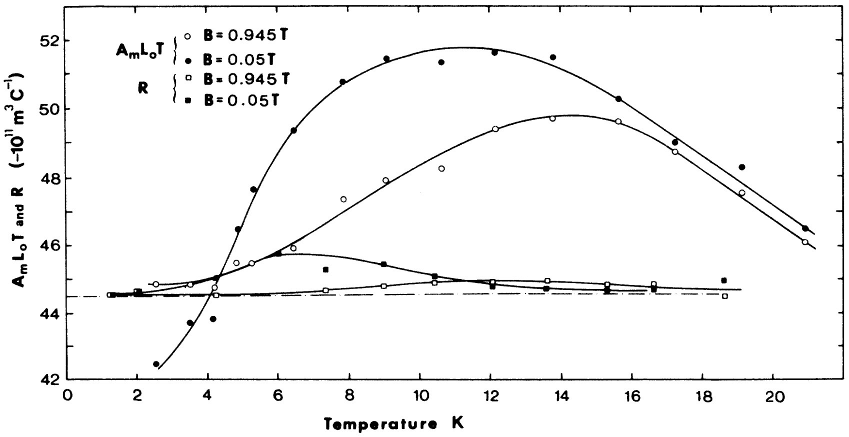

FIG. 6. Results on A_mL_0T and R for K9 taken at fields of 0.945 and 0.05 T. For $B=0.05$, $\omega_c\tau \simeq 0.3$ (using $R\sigma B=\omega_c\tau$). For $B=0.945$ T, $\omega_c\tau \simeq 6$ at 4 K and $\omega_c\tau_\sigma \simeq 1$ at roughly 15 K; notice that the peak in A_mL_0T is very near the temperature at which $\omega_c\tau_\sigma \sim 1$ for the high-field data.

for K, $(ne)^{-1}=4.450\times10^{-10}$ m^3 C^{-1} at 5 K, the former being determined for the bcc phase; the hcp phase of Na has an almost identical atomic volume and gives $(ne)^{-1}=-2.360\times10^{-10}$ m^3 C^{-1}. In all cases the experimental values tend to the theoretical values at low temperatures, well within the experimental uncertainty due to the thickness measurements; the mean value of R for the all Na samples is -2.33×10^{-10} m^3 C^{-1}, and for the all K samples we obtain -4.44×10^{-10} m^3 C^{-1}.

B. Low Fields and Intermediate Temperatures

At low fields Eq. (3) is no longer valid, and the anisotropy of the electronic scattering becomes significant. The temperature variation of R for Li, Na, K, and two dilute K alloys has been investigated by Alderson and Farrell[2]; they have analyzed their results using an expression, which, for a spherical Fermi surface, can be reduced to

$$R=\frac{1}{ne}\,\frac{\langle\tau^2\rangle}{\langle\tau\rangle^2}\,, \tag{4}$$

where τ is the electronic relaxation time and the brackets signify averaging of the quantity over the Fermi surface. Equation (4) is strictly valid only when the scattering is elastic, and even then only at low fields,[15] i.e., $\omega_c\tau \ll 1$. However, it seems reasonable to assume that it would provide a first approximation when the scattering is not elastic, providing we use τ_σ the relaxation time appropriate to the electrical conductivity, i.e.,

$$R=\frac{1}{ne}\,\frac{\langle\tau_\sigma^2\rangle}{\langle\tau_\sigma\rangle^2}. \tag{5}$$

Hayman and Carbotte[16] have used Eq. (5) to theoretically examine the behavior of R of the alkalis. They show that τ_σ due to phonon scattering in Na and K is reasonably isotropic above about $\Theta_D/2$

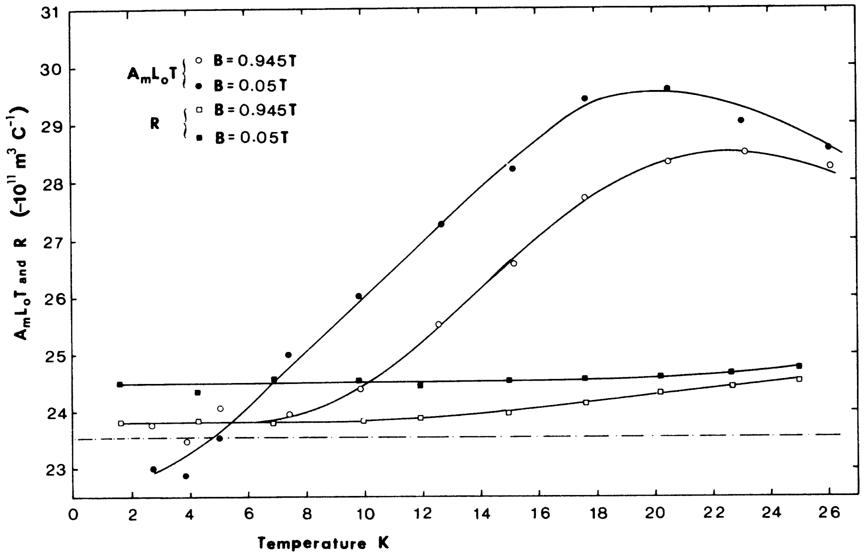

FIG. 7. Results on A_mL_0T and R for Na8 taken at $B=0.945$ and at 0.05 T, using the data of Fig. 5 and other similar data. At $B=0.05$ T, $\omega_c\tau \simeq 0.4$ at 4 K. For $B=0.945$ T, $\omega_c\tau \simeq 4$ at 4 K and $\omega_c\tau_\sigma \simeq 1$ at roughly 22 K; notice that the peak in A_mL_0T is very near the temperature at which $\omega_c\tau_\sigma \sim 1$ for the data taken at the higher field.

(Θ_D being the Debye temperature; about 160 K for Na, 100 K for K), but can be highly anisotropic at low temperatures. Thus at the lowest temperatures (where $\omega_c\tau \gg 1$) and at the highest temperatures (where $\langle\tau_\sigma^2\rangle \simeq \langle\tau_\sigma\rangle^2$) we expect Eq. (3) to be valid, but at intermediate temperatures (where $\langle\tau_\sigma^2\rangle > \langle\tau_\sigma\rangle^2$) R should pass through a maximum. This is exactly the situation found experimentally in both the present work and in that of Alderson and Farrell.[2] Since the magnitude of $\omega_c\tau$ governs the transition from low- to high-field conditions, the exact magnitude of the peak and the temperature at which it occurs will depend on the value of the magnetic field used in the measurement. It would appear that Alderson and Farrell have used Eq. (5) down to the lowest temperatures even though it is quite likely that their samples are in the high-field region (they do not specify the magnetic field, and we assume it is of the order of 1 T). On the basis of the foregoing arguments, it might be expected that R is field dependent at low temperatures. We have investigated this point by measuring R over a wide range of field (0.05–0.95 T), and these results are shown in Figs. 6 and 7. Table I lists values of $\omega_c\tau$ for all the samples and in particular for K9 and Na8, for which the low-field measurements were made. At a magnetic field 0.05 T and a temperature of 4 K, $\omega_c\tau = 0.31$ for K9 and 0.40 for Na8. The field dependence of R is clearly visible, though of relatively small magnitude. The limiting low-field value of R at low temperature is presumably determined by the residual impurities in the samples, which may or may not cause anisotropic electronic scattering. In the case of K9, it appears that the limiting low- and high-field values are the same, suggesting isotropic scattering, but the Na8 sample seems to have different limiting values, suggesting anisotropic scattering.

The temperature dependence of A_mL_0T is not so easily accounted for. At intermediate temperatures we do not expect Eq. (1) to be valid, even if we ignore the effects of λ_g. The reason is that any scattering anisotropy can influence R and A to different extents in general. Consider first the elastic-scattering low-field limit and spherical Fermi surface, where we have an equation for A exactly analogous to Eq. (4), i.e.,

$$AL_0T = \frac{1}{ne}\frac{\langle\tau^2\rangle}{\langle\tau\rangle^2}\,, \tag{6}$$

and in this limit R is identical with AL_0T. Again it seems reasonable to assume that we can apply Eq. (6) as a first approximation when the scattering is inelastic (as we did in the case of the Hall effect), providing we use the electronic relaxation time τ_λ appropriate to the *thermal* resistivity; this should be contrasted with the expression for R, where τ_σ is the appropriate quantity in Eq. (5). Including Eq. (2) in Eq. (6) we have

$$A_mL_0T = \frac{1}{ne}\frac{\langle\tau_\lambda^2\rangle}{\langle\tau_\lambda\rangle^2}\left(\frac{\lambda_e}{\lambda}\right)^2. \tag{7}$$

The point to emphasize is that, providing τ_λ and τ_σ have the same scattering anisotropy and if λ_g can be ignored, we return once more to Eq. (1) *even when the electronic scattering is inelastic*. Sondheimer[17] has shown that Eq. (1) is indeed satisfied for a spherical Fermi surface if the scattering is inelastic, though his proof is based on no scattering anisotropy. This of course is contrary to the behavior of the electrical and thermal conductivities, where an equation analogous to Eq. (1) does not hold when the scattering is inelastic. (The Appendix gives a simple physical picture of why Eq. (1) does hold for isotropic inelastic scattering.)

We have attempted to allow for the factor $(\lambda_e/\lambda)^2$ by using the theoretical estimates[18] for λ_g, i.e., $\lambda_g = 240/T$ W m^{-1}K^{-1} for Na and $\lambda_g = 100/T$ W m^{-1}K^{-1} for K; the corrected data are plotted on Figs. 2 and 4. This correction produces an estimate of the quantity AL_0T (i.e., the Righi-Leduc coefficient if the lattice conductivity were negligible) rather than A_mL_0T. Thus the corrected data, labeled A, should be analyzed using the expression

$$AL_0T = \frac{1}{ne}\frac{\langle\tau_\lambda^2\rangle}{\langle\tau_\lambda\rangle^2}\,. \tag{8}$$

As in the case of R, we have $\langle\tau_\lambda^2\rangle \geq \langle\tau_\lambda\rangle^2$ with the equality applicable only to the isotropic scattering case, and thus we expect that $(ne)^{-1}$ is the minimum absolute value of AL_0T. The data of Figs. 2 and 4 are contrary to this expectation at higher temperatures. This suggests that λ_g is possibly larger than the theoretical estimates by a factor of perhaps 2–3 for both Na and K. However, we should point out that we obtained similar results for the noble metals, reported in I, particularly in the case of Ag. Furthermore the theoretical estimates of λ_g for the cubic metals have so far been consistently *higher* than the available experimental values by roughly a factor of 2.[19] Thus on the basis of the available evidence one might expect the correction for λ_g to be less than we have applied rather than more, as we would need for experimental and theoretical agreement.

At lower temperatures we see that AL_0T (and A_mL_0T, for which the behavior is qualitatively the same) shows a relatively large peak for both Na and K. It is significant that the position of this peak occurs very close to the temperature at which $\omega_c\tau_\sigma \sim 1$; for $B = 1$T these temperatures are approximately 15 K for K and 22 K for Na, and have been obtained using $\omega_c\tau_\sigma = R\sigma B$. Presumably a more appropriate quantity would be $\omega_c\tau_\lambda$, but we expect τ_σ

and τ_λ to be within a factor of 2 of each other, and the rapid temperature dependence of both τ_λ and τ_σ will cause only slight shifts in the temperatures so estimated. As we have already mentioned, the measurements were made at fields of 0.525 T and 0.945 T. It was noticed that at the lower field the position of the peak was 1–2 K lower in temperature. We have investigated the field effects more fully by taking measurements on $A_m L_0 T$ over a wide range of field down to approximately 0.05 T (500 G). The results of these experiments are shown in Figs. 5–7. The effect of the decrease in field is far more significant for $A_m L_0 T$ than for R, and as we should expect the effects are only noticeable below those temperatures for which $\omega_c \tau \sim 1$. It is worth mentioning that the position of the peak in A at low fields scales quite accurately with Θ_D for Na and K.

If we return to Eq. (8) we see that any increase in the absolute magnitude of $A L_0 T$ must be explained according to our analysis by the factor $\langle \tau_\lambda^2 \rangle / \langle \tau_\lambda \rangle^2$, and we should be comparing this expression with our data taken at lowest field (ideally as $B \rightarrow 0$, which should be reasonably well satisfied). We feel that it is unlikely that this factor really is the cause of the large peaks in $A_m L_0 T$ that we observe. First consider the case of R, where any difference between $(ne)^{-1}$ and R is to be associated with $\langle \tau_\sigma^2 \rangle / \langle \tau_\sigma \rangle^2$. The measurements show that this factor is quite small, being a maximum of about 1.03 for both K and Na. To explain the $A_m L_0 T$ data we require that $\langle \tau_\lambda^2 \rangle / \langle \tau_\lambda \rangle^2$ be a maximum of 1.19 for K and 1.23 for Na, which would seem to be unreasonable. The only difference between τ_σ and τ_λ is the geometric factor involving the change in electron velocity during the scattering event that must be included in the former, but since this depends upon the variation of the phonon velocity in the various directions as well as the relative importance of N and U processes, one cannot be sure, without a full calculation, of the relative magnitudes of the scattering anisotropies of τ_λ and τ_σ.

It is worth evaluating other possible causes for the peak in $A_m L_0 T$. Chambers[20] has given the results of the application of the two-band model to A, which we might attempt to apply to the alkalis by associating the two bands with two groups of electrons with different properties. His Eq. (30) can be written,[21] for the case of low fields,

$$A = \frac{\lambda_1^2 A_1 + \lambda_2^2 A_2}{\lambda^2} + \frac{T}{\lambda^2} \left(\frac{\sigma_1 \sigma_2 \Delta\epsilon}{\sigma} \right)^2 \times [R_1 + R_2 + 2\Delta\epsilon\, \Delta B_{\mathrm{EN}}^{i} (\sigma_1 + \sigma_2)], \qquad (9)$$

where the subscripts 1 and 2 refer to the two bands, $\Delta\epsilon$ is the difference in thermopower between the two bands, and ΔB_{EN}^{i} is the difference in the Nerst-Ettinghausen coefficient between the two bands. The first term involving A_1 and A_2 is essentially equivalent to Eq. (8), for a spherical Fermi surface (see also I). Thus if the only difference between the two groups of electrons is their relaxation time τ_λ, then $A_1 = A_2$, and if the areas of the Fermi surface appropriate to the two groups are S_1 and S_2, this first term reduces to $A_1 (S_1 \tau_1^2 + S_2 \tau_2^2) / (S_1 \tau_1 + S_2 \tau_2)^2$, which is just a particular case of Eq. (8). The origin of the second term in Eq. (9) can be traced to the experimental conditions that require that there be no electric current density in the sample, i.e., $J_x = 0 = J_y$. However, this does not mean that the currents associated with the two bands must be individually zero, only that $J_{1x} + J_{2x} = 0 = J_{1y} + J_{2y}$. It is straightforward to show, and this was pointed out by Chambers, that the magnitude of the second term is very small, being less than or of the order of 0.1% of the first term. Thus Eq. (8) throws no new light on the problem.

Another result of possible significance is that due to Tsuji.[9] Tsuji investigated the effects of the nonequilibrium distribution of phonons due to the temperature gradient on R and A in particular.[21] He did not investigate the effects of the anisotropy of the electronic relaxation time, but he did include in his calculation the effect of a distorted Fermi surface having cubic symmetry. It was found that the resulting phonon drag effects do not influence R, but can have a potentially large effect on A. His result shows, at low temperatures, that the magnitude of the phonon-drag term A_{ph} compared to the ordinary term A should be

$$\frac{A_{\mathrm{ph}}}{A} \sim \frac{\zeta}{kT} \left(\frac{T}{\Theta_D} \right)^3 ,$$

where ζ is the chemical potential and Θ_D the Debye temperature. For $(T/\Theta_D) \sim 0.1$ this ratio can be of the order of unity. In the case of a spherical Fermi surface the phonon drag term reduces to zero in this theory. However, Tsuji used a Debye approximation for the phonon spectrum, and it may be that the strong anisotropy of the phonon spectrum of the alkalis can modify this result considerably. It remains for such a modified calculation to be attempted.

Whether the peak in A is due to phonon drag or to the anisotropy of τ_λ, or both, one would expect that high fields would eliminate the effect. Both of the possible mechanisms depend on anisotropy, and the effect of a high field would be to average out the properties of the electrons over the Fermi surface by allowing the electrons to sample all the states on an orbit over the Fermi surface lying in plane perpendicular to the magnetic field. Thus the experimental reduction of the peak at high fields is consistent with both the above explanations. In view of the uncertainty of the origin of the peaks in A, we feel it would be unprofitable to discuss the observed effect of the phase change of Na on the results.

IV. CONCLUSIONS

We find the low-temperature high-field limiting values of A and R to be as expected for both K and Na. However, although R varies only weakly with temperature, AL_0T shows a pronounced low-temperature peak for which there appears to be two possible explanations.

(i) The anisotropy of the thermal relaxation time is much more pronounced than that of the relaxation time appropriate to the electrical conductivity. We feel the observed magnitudes make this unlikely, but cannot clearly rule out the possibility.

(ii) Phonon drag is responsible for the peak. There are no calculations available that can be considered appropriate to the alkalis, i.e., taking into account the anisotropic phonon spectrum and retaining spherical energy surfaces.

At higher temperatures the magnitude of A_mL_0T falls well below that of R, possibly suggesting that the lattice thermal conductivity is higher than expected. It is clear that more theoretical work is required on the Righi-Leduc effect before the situation can be clarified.

APPENDIX

At first sight it might be surprising that Eq. (1) of the text holds (at least when the energy surfaces are spherical) when the scattering is inelastic, in contrast to the case of the electronic and thermal conductivities. The following simple picture shows why this is physically the case.

In the case of the Righi-Leduc effect, the longitudinal heat current U_x produces a deviation from equilibrium, as is shown schematically in Fig. 8. Under the influence of a magnetic field in the z direction, the distribution is rotated through an angle $\omega_c\tau_\lambda$ to give a transverse heat current in the y direction of $U_y = \omega_c\tau_\lambda U_x$ (if $\omega_c\tau_\lambda \ll 1$). Experimentally it is arranged that $U_y = 0$, so that a transverse temperature gradient is established of

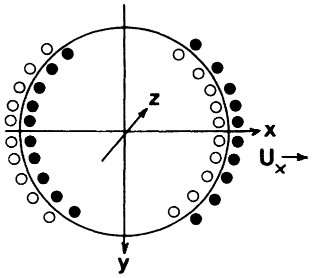

FIG. 8. Deviation from equilibrium for the electrons under the influence of a temperature gradient in the $-x$ direction. ● and ○ denote an excess or deficiency of electrons, as compared to the equilibrium distribution.

$$\frac{\partial T}{\partial y} = -\frac{U_y}{\lambda};$$

using $\lambda = \pi^2k^2nT\tau_\lambda/3m$ appropriate to a spherical Fermi surface (and ignoring λ_g), we finally have

$$A = -\frac{\partial T/\partial y}{BU_x} = \frac{1}{L_0Tne} = \frac{R}{L_0T}.$$

An analogous argument holds for the Hall effect, but notice that in the case of the Hall effect, the angle through which the distribution is rotated is $\omega_c\tau_\sigma$, which might be much greater than $\omega_c\tau_\lambda$ in the inelastic scattering regime; thus the Wiedemann-Franz law $AL_0T = R$ does not imply, as has been suggested,[22] that the Hall and Righi-Leduc angles are equal, and in fact they may be totally different.

Using the above analysis, it is easy to see how the factor $(\lambda_e/\lambda)^2$ arises in Eq. (2); thus the electronic heat current is $(\lambda_e/\lambda)U_x$ and the transverse temperature gradient is really $-U_y/\lambda$ or $-(U_y/\lambda_e) \times (\lambda_e/\lambda)$. This last result does not depend on either the spherical-band approximation or elastic scattering.

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