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Temperature dependence of the electrical resistivity of potassium films

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A study of the temperature dependence of the electrical resistivity of rolled potassium films is reported for the temperature range 4.2–17.4 K. The thickness of the samples was between 3.4 and 69.0 μm, which is comparable to the mean free path of the electronic motion for the pure bulk potassium. This implies the appearance of size effects in the electrical resistivity. The study covered the region for which the temperature dependence of the electrical resistivity is due to the electron-phonon *umklapp* scattering process, and can be described by the expression $(T/\Theta)^n \exp(-\Theta/T)$. The exponent n of the prefactor in the electron-phonon scattering term showed a linear increase as a function of the reciprocal of the film thickness, $1/d$, when the parameter Θ was kept constant. A corresponding linear increase in Θ as a function of $1/d$ was observed when the fitting was done with a constant value of n. From the comparison with the available theoretical analysis for the bulk case, we find that the interrelation between the n and Θ parameters is readily established for thin potassium films.

I. INTRODUCTION

The electrical resistivity of metals and especially its variation with temperature has been a very important transport property of materials and has been studied widely both theoretically and experimentally.[1–4] A special interest has been given to the study of the electrical resistivity of potassium because it has an almost spherical Fermi surface and a cubic lattice. Several pioneering studies of the temperature dependence of the electrical resistivity of potassium have been completed by Dugdale and Gugan (4–300 K),[5] Ekin and Maxfield (4–20 K),[6] and Gugan (below 4 K).[7] It was shown that, in the region between 2 and 5 K, the electron-phonon *umklapp* scattering strongly predominated the electron-electron scattering.[8,9] Several attempts have been made to study the significance of the electron-electron scattering mechanism below 2 K, with high-precision measurements by Pratt,[10] Yu *et al.*,[11] Zhao *et al.*,[12] Rowlands *et al.*,[13] Levy *et al.*,[14] and H. van Kempen *et al.*[8] These experiments encouraged theoretical investigations of the electrical resistivity of potassium below 2 K (see Refs. 1–3 for a discussion of the theoretical developments).

The majority of the previous experimental work[5–8,10–14] was performed on thick-wire samples with diameters between 0.7 and 3 mm, except for several thin specimens studied at low temperatures by Yu *et al.* and Zhao *et al.* at Michigan State University.[11,12]

Although an explanation of some experimental anomalies in the resistivity below 2 K was attempted in terms of size-effect phenomena,[12,15] i.e., due to the closeness of one of the sample dimensions to the electronic mean free path (MFP),[16] it appears that the size effects in the higher-temperature region, where the scattering by the electron-phonon *umklapp* process is dominant, have remained unstudied in potassium both theoretically and experimentally.

It has been shown by Sambles and co-workers[17] that some data for the temperature dependence of thin films and wires can be interpreted quite well in terms of a phenomenological model of size effects which includes the influence of the reduced sample thickness upon an implicit background-scattering mechanism. The important model variable for this type of model is $\kappa = d/\lambda$, where d and λ are the sample thickness and the MFP of electrons in the bulk material, respectively. Tellier and Tosser[18] noted that the majority of the size-effect models, which study the temperature dependence of the resistivity, are not readily applicable to the low-temperature case because of their usual assumption of a linear dependence of the electronic MFP on temperature. This assumption restricts these models to the high-temperature regime, where the electron-phonon interaction is dominant and the scattering amplitude is proportional to the mean-square thermal fluctuations of the lattice.[19] To our knowledge there is no available theoretical treatment of the temperature-dependent electrical resistivity of size effects in thin films by solving the coupled Boltzmann transport equations for electrons and phonons under the restriction of surface boundary conditions applied to both the electrons and phonons. Usually, classical size effects are treated in a "rigid-boundary model," i.e., not including boundary relaxation effects, such as surface phonons and a possible excitation of low-energy slab modes via the interaction of electrons with highly inhomogeneous surface potentials.

In this paper we present a study of the temperature dependence of the electrical resistivity of thin potassium films (3.4–69.0 μm) in the temperature range 4.2–17.4 K. All the samples were checked for the reproducibility of the size effects in magnetoresistance[20] in order to be sure that the surface scattering is large enough to be observable in the resistivity measurements.

II. EXPERIMENTAL PROCEDURE

The sample preparation of the potassium films followed the procedure which is described fully in the cited report of magnetoresistivity measurements (first of Ref. 20).

We used a calibrated carbon-glass–resistance thermometer, model CGR-1, Lake Shore Cryotronics, Inc., for the temperature reading inside the experimental chamber. Temperature was increased by means of a Constantan wire heater, which was packed into a small sealed brass cylinder filled with silicon vacuum grease. The brass cylinder was in direct contact with a bulk piece of potassium with a mass of 0.5 g which served as a heat reservoir for the potassium-film sample. The thermometer was placed in direct contact with the potassium metal. We thus provided a very good metal-to-metal thermal contact between the thermometer, heater, and potassium-film sample without having any trouble from electrical shorts.

The in-plane dimensions of the Kel-F substrate, which supported the film sample with length L and width W were the same for all our samples with $L=4.83$ cm and $W=0.40$ cm. The sample thickness was determined 3 h after the film preparation from the room-temperature, four-probe resistance (see Table I) using the bulk resistivity of pure potassium.

Our chamber provided us with a temperature stability to within 12 and 100 mK at the lower and upper limits of the 4.2–17.4-K temperature region of the study, respectively. The potential reading was detected by a Keithley nanovoltmeter, model 149, with a typical standard deviation of the voltage data of 10 nV. The typical reproducibility of the resistance measurements at the middle of the temperature range was 0.7% during one experiment. The data for each sample were collected in at least three (mostly four) different experimental runs, with warming up to 80 K and cooling back to 4 K between consecutive runs. Some deviations of the measurements at the same temperature in different runs exceeded 0.7%. This we attribute to the possibility of rearrangement of vacancies and interstitials in potassium at $T>20$ K.[21]

III. RESULTS AND DISCUSSION

The usual method of data analysis for bulk specimens relies on the assumption that there are independent scattering processes which contribute to the electrical resistivity. This is known as Matthiessen's rule.[4] For the low-temperature regime, the temperature dependence of the resistivity, $\rho(T)$, is described by[19]

$$\rho(T)=\rho(0)+AT^5+BT^2+C(T/\Theta)^n\exp(-\Theta/T)\ , \tag{1}$$

where $\rho(0)$ is the residual resistivity, which does not depend on T and is due to scattering of electrons from impurities and other imperfections in the crystal. The AT^5 term corresponds to the low-temperature limiting dependence of the resistivity from normal electron-phonon scattering. This can be very small if the phonons drift with the electrons (the so-called phonon-drag effect). The third term is due to electron-electron scattering and contributes at all temperatures, although it is very small for potassium at temperatures above 1.5–2 K.[12,22] The high-accuracy measurements of van Kempen *et al.*[8] show that the AT^5 term from the normal electron-phonon scattering term is insignificant even in the temperature range 1–4 K. They also provide evidence of the unimportance of the electron-electron BT^2 scattering term for $T>2$ K and support the picture of electron-phonon scattering under strong phonon-drag conditions of the *umklapp* process (first paper among those listed in Ref. 8). The most significant term in Eq. (1) in our temperature region is the fourth term, which describes the electron-phonon *umklapp* scattering, Θ being the characteristic activation energy for the phonons participating in the scattering *via* an *umklapp* process. The bulk value of Θ was found to be 19.9 and 23 K in two different experiments.[6,8] The scattering term which is due to the *umklapp* electron-phonon process is the main concern of the present study, since it is responsible for the rapid increase in the electrical resistivity of bulk potassium in the region 4–20 K.[6–8] Thus, for a bulk sample the simplified form of Eq. (1) is

$$\rho(T)=\rho(0)+C(T/\Theta)^n\exp(-\Theta/T),\quad 4<T<20\text{ K}\ . \tag{2}$$

Let us note that in Eq. (2) we exclude from our consideration the contribution from the normal electron-phonon scattering relative to that provided by the *umklapp* process. Ekin and Maxfield,[6] in their study of samples that show no size effect, found that in the region 2.5–25 K the dominant contribution to the electrical resistivity is due to the *umklapp* scattering mechanism. They also calculated the contribution to the resistivity by the normal processes (see Table III in Ref. 6) using several different forms of the pseudopotential. Examining their Table III, one finds that the theoretical estimation for changes of the normal component is from 15%, 27%, and 33% of the total resistivity at 4 K to 19%, 29%, and 42% at $T=18$ K for the Bardeen, Lower, Lee–Falicov, and Ashcroft pseudopotentials, respectively. Leavens and Laubitz[23] show that under strong phonon-drag regime (see Fig. 4 in Ref. 23) the normal component in the region 4–18 K is reduced by a factor of 2–3 compared to the calculations of Ekin and Maxfield,[6] but still exhibits an almost linear increase from 15% to 23% in this temperature interval. Since there is no theoretical treatment of the contribution of the normal processes to the temperature-dependent resistivity of potassium films, which show size-effect-related behavior,[20] we make the *ad hoc* assumption that the normal processes contribute a linear slowly varying background to the dominant temperature dependence due to the electron-phonon *umklapp* scattering mechanism. In Fig. 1 we show the experimental data of the present study of the temperature dependence of the electrical resistivity of thin films together with its typical behavior that has been observed for bulk samples by Ekin and Maxfield.[6] One observes that the data support a strong nonlinear increase, which is a characteristic feature of the *umklapp* processes. A clear manifestation of the size effects is evident from the increase of the resistivity as the sample thickness d is decreased. However, since the temperature dependence of our samples qualitatively resembles the behavior from bulk specimens, we apply the form of Eq. (2) to the analysis of the data from the thin films in order to study the implications of the reduced thickness on the parameters n and Θ of Eq. (2). We write, therefore, for the

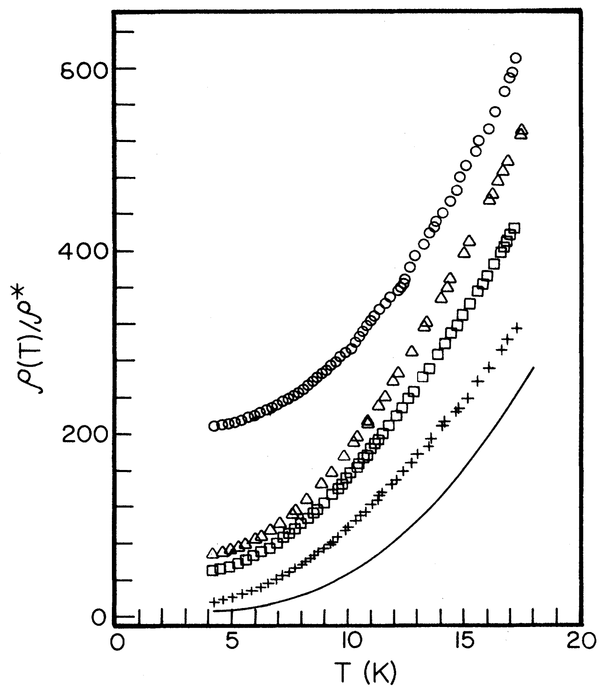

FIG. 1. Temperature dependence of the electrical resistivity of samples 1–4 in units of the ideal bulk resistivity of potassium, $\rho^*=2.94\times10^{-10}$ Ω cm, at $T=4.25$ K (see Table II in Ref. 6). The solid line gives the data from Table II of Ref. 6 according to $\rho=\rho_0+\Delta\rho(T)$. $\square$, $\circ$, $+$, and $\triangle$ correspond to 1, 2, 3, and 4 of the present study, respectively.

size-effect case,

$$\rho_S(T)=\rho_S(0)+C_S(T/\Theta)^n\exp(-\Theta/T), \quad 4<T<20 \text{ K} \tag{3}$$

where $\rho_S(0)$ and C_S are allowed to vary from sample to sample.

First, we have to eliminate the sample-dependent quantities $\rho_S(0)$ and C_S in Eq. (3). Using the measured value of $\rho_S(T_0)$, at the temperature $T_0=4.2$ K, we eliminate the unknown $\rho_S(0)$ term and write, with the aid of Eq. (3), the following:

$$\begin{aligned}F(T)&=[\rho(T)-\rho(T_0)]/[\rho(T_m)-\rho(T_0)]\\&=[P(T)-P(T_0)]/[P(T_m)-P(T_0)]\ ,\end{aligned} \tag{4}$$

where T_m is the upper limit of the temperature region for each sample [we omit the subscript S in $\rho(T)$ for clarity] and

$$P(T)=(T/\Theta)^n\exp(-\Theta/T) \tag{5}$$

is assumed to satisfy Matthiessen's rule for the bulk sample. Then, since the right-hand side of Eq. (4) is sample independent, the quantity $F(T)$ derived from the resistance measurements should also show sample-independent temperature behavior. It is clear that this form of $F(T)$ provides a systematic uncertainty of about 10% to the analysis of our data, since we neglect the linear increase in the relative contribution of the normal processes in the 4.2–17.4-K temperature range. We note also that, according to Kaveh *et al.*,[24] the parameters n and Θ in Eq. (5) should be chosen differently (i.e., they should have a temperature dependence) when the fitting to the data is performed over a wide range of temperature. In Fig. 2 we plot $F(T)$ for the thickest and the thinnest samples, 3 and 2, respectively. The $F(T)$ of samples 1 and 4 fall inside the limits provided by the curves presented in Fig. 2. One sees that the data presentation in the form of $F(T)$ results in very similar curves for the temperature dependence of all our samples, which have their thickness in the range between 3.4 and 69.0 μm, except for a small thickness-dependent deviation from the strict formal Matthiessen's rule.

Our first attempt was to try to fit our data for the thickest sample, 3, with the pair $n=1$, $\Theta=20$ K, which was found by van Kempen *et al.*[8] for bulk samples. However, this was not satisfactory. Thus, in Fig. 3(a) we present the best fit to the data of sample 3, which are plotted in the form of $F(T)$, defined by the left-hand side of Eq. (4), to the expression given by the right-hand side in terms of $P(T)$. It is clear that the most sensitive region of the experimental and interpolation curves is the middle of the studied temperature interval, because of the fixed values of 0 and 1 at $T=T_0$ and $T=T_m$, respectively.

In Fig. 3(b) we compare our fitted curve with $n=1.00$

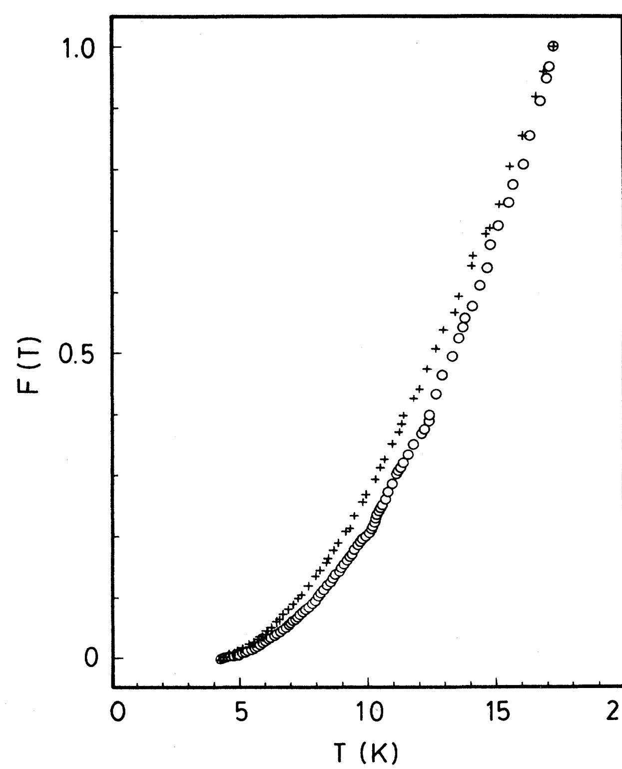

FIG. 2. Plot of the quantity $F(T)$, defined by the left-hand side of Eq. (5); $+$ and $\circ$ correspond to the thickest and thinnest samples, 3 and 2, respectively.

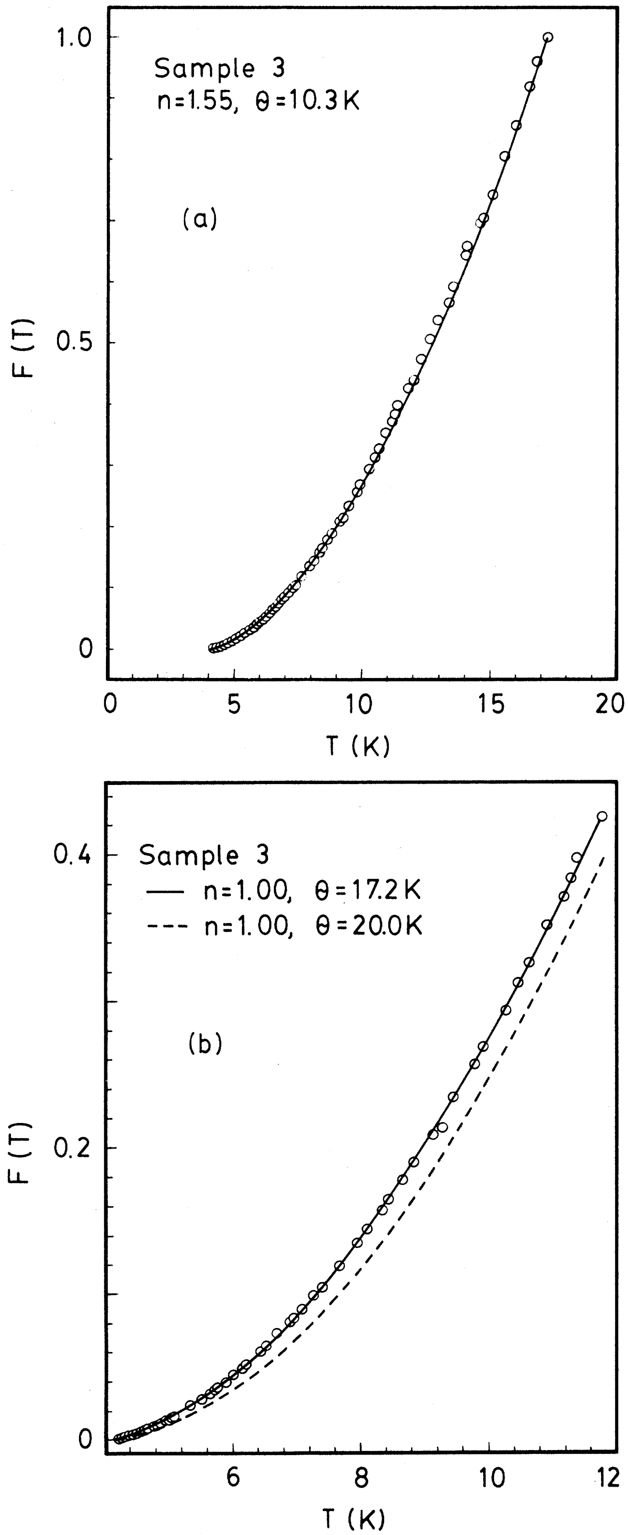

FIG. 3. (a) Fit to $F(T)$, defined by the left-hand side of Eq. (5), using the right-hand side of the expression, which is expressed via $P(T)$ of Eq. (6), with $n=1.55$ and $\Theta=10.3$ K. (b) Comparison between the best fit to our data by means of the same procedure in (a), but with the pair $n=1.00$, $\Theta=17.2$ K (solid line) and the line, suggested by van Kempen *et al.* (Ref. 8) for the bulk case $n=1$, $\Theta=20$ K (dashed line).

and $\Theta=17.2$ K to that found for the bulk case: $n=1.00$, $\Theta=20.0$ K.[8] One sees that there is a clear difference between the results for the bulk and the size-effect cases. Keeping $n=1$, we found that the value of Θ showed a clear decrease with an increase of the sample thickness and did not approach the $\Theta=20$ K bulk limit. This is illustrated in Fig. 4, where we plot Θ, which was derived from the best fits to our data, versus the reciprocal of the dimensionless parameter $\kappa=d/\lambda$ ($\lambda=66$ μm). We note that the intersection point gives $\Theta=16.8\pm0.2$ K, which is quite close to the value of 17.6 K derived by Frobose[27] for the bulk limit. However, in spite of the series of the simplifying assumptions that are mentioned above in connection with Eqs. (2) and (3), the Θ value extrapolated to the bulk limit, $\Theta=16.8$ K, is considered to be in good agreement with the existing bulk data, especially since the latter were derived for a lower- (1–6 K) temperature region.[6–8,24–27]

According to Kaveh *et al.*,[24] for bulk potassium in the 3–6-K region one might try the pair $n=\frac{7}{2}$ and $\Theta=8$ and the (n,Θ) pair should be chosen differently for each limited temperature region. However, our data could be fitted, within experimental accuracy, with the same (n,Θ) pair over the whole temperature range 4.2–17.4 K. In order to check the convergence of the size-effect values of n to the bulk case, $n=\frac{7}{2}$, we kept Θ constant at a value that is slightly different from that of Kaveh *et al.*[24] We fixed $\Theta=10.3$ K, which corresponds to the energy of the $T2$ mode in the [110] direction,[27,28] and varied the value of n. The result is presented in Fig. 5, where the bulk limit for n is $n=1.52\pm0.02$ instead of $n=\frac{7}{2}$. Even when we use, for fitting, the $\Theta=8$ K suggested by Kaveh *et al.*,[24] the bulk limit for n increase to $n=1.68$, which is still much less than $\frac{7}{2}$. The difference between the extrapolated value of n, $n=1.5$, and its theoretical counterpart, $n=\frac{7}{2}$, is probably due to our oversimplifying assumptions [see Eqs. (2) and (3)]; therefore we stress here only that n is almost a linear function of the reciprocal of

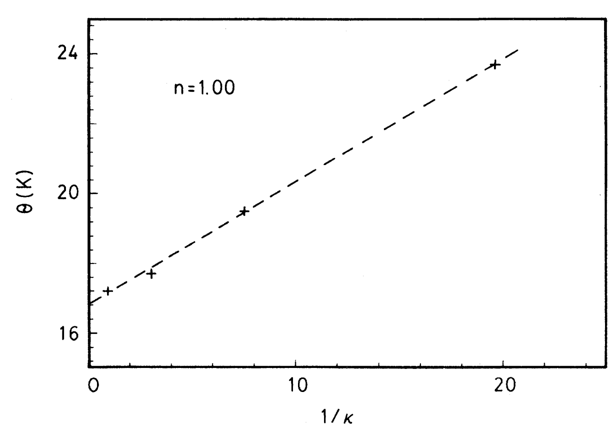

FIG. 4. Linear increase of Θ (fixed $n=1.00$) as a function of the reciprocal of the dimensionless parameter $\kappa=d/\lambda$, where d is the film thickness and $\lambda=66$ μm is the electronic mean free path.

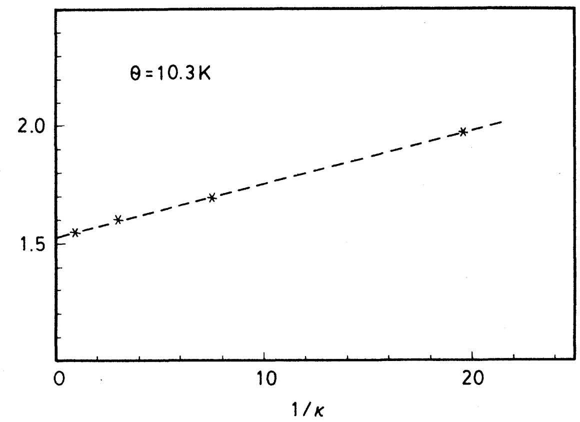

FIG. 5. Linear increase of n (fixed $\Theta = 10.3$ K) as a function of the reciprocal of the dimensionless parameter $\kappa = d/\lambda$, where d is the film thickness and $\lambda = 66$ μm is the electronic mean free path.

the sample thickness, d. In Table I we present the n and Θ values together with some other useful parameters of our samples 1–4.

Within the accuracy of our measurements, we find that alternative (n,Θ) pairs fit the experimental data equally well for each sample over the whole range of temperature, 4.2–17.4 K. Keeping Θ (or n) fixed and varying the values of n (or Θ) in the fitting to the same set of data for each sample, we observe that the increase (decrease) of Θ (n) corresponds to a decrease (increase) in n (Θ), according to $\Delta\Theta/\Delta n = -12.6$, -12.3, -13.3, and -13.8 K for samples 3, 1, 4, and 2, respectively, where the listing is in the order of decreasing sample thickness (see Table I). Taking the average of $\Delta\Theta/\Delta n$ over all the samples,

$$\Delta\Theta/\Delta n = -13.0 \text{ K}, \quad 4.2 < T < 17.4 \text{ K} . \tag{6}$$

The possibility of fitting the same set of data with quite different (n,Θ) pairs was mentioned first by Gugan[7] in his study of the electrical resistivity of potassium; he found that within the precision of his study the data could be fitted almost equally well by different n and Θ, provided that $\Theta = 23.6 - 2.8n$ K. This point was clarified by Kaveh, Leavens, and Wiser,[24] who also explained the apparent discrepancy between the observations and the theoretical predictions for the (n,Θ) pair. They especially noted that in the region $T = 3-6$ K there exists the possibility of fitting experimental data with quite different (n,Θ) pairs, provided that

$$\Delta\Theta/\Delta n = -3 \text{ K}, \quad 3 < T < 6 \text{ K} . \tag{7}$$

This says that for a limited temperature region an increase in n implies a decrease of Θ, for one set of data. Unfortunately, the analysis[24] of the interrelation between the n and Θ parameters is limited to the temperature region $T < 5-6$ K.

On the grounds of this finding, we propose that the analysis of Kaveh *et al.*[24] can be extended from its region of validity (3–6 K) to that studied here for the case of thin potassium films.

Let us note that the increase of n (fixed Θ) and Θ (fixed n) with increasing $1/d$ still requires more extensive study. The question—will this almost linear increase in n or Θ versus $1/d$ saturate at the limit of very thin films?—remains open.

IV. SUMMARY AND CONCLUSION

All the samples studied in this experiment were checked first for reproducibility of size effects in magnetoresistance in order to be sure that the surface scattering was large enough to be observable in the resistivity measurements.

We found that the temperature dependence of the electrical resistivity in thin samples in the $T = 4.2-17.4$-K region, where the electron-phonon *umklapp* scattering is dominant, can be described in the case of thin films by the expression in the form previously reported for bulk specimens, for a low-temperature region.

The temperature dependence of the electrical resistivity of potassium films can be presented in a form of Matthiessen's rule with quite small deviations that are function of the film thickness.

From the study of the thickness dependence of these deviations, we found that for a fixed value of n in the term $(T/\Theta)^n \exp(-\Theta/T)$ the corresponding Θ values show a linear increase with increasing $1/d$, the reciprocal of the sample thickness. We also observed the linear increase of n with increase of $1/d$ for Θ fixed at the

TABLE I. Various parameters of the potassium-film samples studied in this work. κ is the film thickness-to-mean-free-path ratio, i.e., $\kappa = d/\lambda$, $\lambda = 66$ μm; R_S is the residual resistivity ratio for film samples [see Eq. (6) for the definition of n and Θ].

Sample no.	R (300 K) (mΩ)	R_S	d (μm)	κ	n [a]	Θ [b] (K)
1	39.38	485	21.8	0.330	1.60	17.7
2	254.5	117	3.4	0.051	1.97	23.7
3	12.42	1392	69.0	1.045	1.55	17.2
4	97.55	359	8.8	0.133	1.69	19.5

[a] n was found for $\Theta = 10.3$ K.
[b] Θ was found for $n = 1$.

theoretical estimate of 10.3 K.

We found that the interrelationship between the n and Θ values predicted for bulk samples at lower temperatures holds for the case of thin films for the temperature range 4.2–17.4 K.

In spite of some oversimplifications used in the course of data analysis, the value for the activation energy Θ extrapolated to the bulk limit agrees satisfactorily with those reported by other authors for thicker samples.

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