MEASUREMENT OF THE ELECTRON MEAN FREE PATH IN POTASSIUM BY MEANS OF THE RADIO FREQUENCY SIZE EFFECT

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Submitted November 20, 1969

Zh. Eksp. Teor. Fiz. 56, 1232–1241 (April, 1969)

The sensitivity of the method with respect to changes in the mean free path l can be increased by increasing the sample thickness d. The behavior of the size effect line for $l < d$ requires an additional investigation. The change in the line shape due to an almost tenfold change in the sample thickness showed that for a relatively small mean free path d the relation between the line shape and field distribution in the skin layer is different from that for $l \gg d$. Some specific conditions (formation of lines by electrons moving along trajectories of various length) are found, which lead to a particularly strong dependence of the line shape on temperature. The temperature-dependent part of the reciprocal path, which defines the line amplitudes, is measured. It is proportional to T^3. In this connection the influence of the collision site on the electron trajectory and the direction of the phonon momentum on the efficiency of electron-phonon collisions is discussed. The residual resistance, whose temperature-dependent part is proportional to T^6, was measured for the same samples by the rotating magnetic field technique.

INTRODUCTION

INTEREST in the measurements of the anisotropy of the mean free path l of the electrons on the Fermi surface has stimulated the development of new methods for measuring l on the basis of effects heretofore used mainly for the study of the energy spectrum. With respect to the radio-frequency size effect (SE), this means a changeover from the study of the locations of the SE lines to a measurement of their amplitudes. Such measurements were already made for tin[1,2], indium[3], gallium[4], cadmium[5], and bismuth[6]. In this paper we investigate certain questions connected with the developement of a method for measuring l of potassium on the basis of the SE. Potassium was chosen because its Fermi surface is, within 0.1%, a sphere of radius $p_F = 0.813 \times 10^{-19}$ g-cm/sec[7], and the SE on potassium was itself investigated in sufficient detail[8,9].

The most convenient for the measurement of l are those SE which admits of only a single passage of the electrons through the "receiving" skin layer, for example the SE at the limiting point. The amplitude A of the lines of such SE is proportional to the number of electrons traversing without scattering the path λ from one side of the plate to the other:

$$A \sim e^{-\lambda/l} = e^{-\alpha d/l} \tag{1}$$

(d is the thickness of the sample, and the proportionality coefficient α can be readily determined from geometrical considerations in each concrete case—see below). As seen from (1), to increase the sensitivity of A to changes of l it is necessary to work in the region

$$l < \lambda, \tag{2}$$

although this leads to a very strong decrease of the absolute value of A. The condition (2) makes it possible to apply formula (1) also to the SE on the central cross section. Although in principle a multiple passage of the electron through the skin layer is possible in this SE, and the amplitude should be determined by an infinite sum of terms of the type (1), nevertheless, owing to the smallness of the free path, only half of the trajectory loops make a contribution to the effect (in the case of a spherical Fermi surface $\alpha = \pi/2$).

In all the prior investigations of the SE line shape, the tendency was to satisfy the inverse condition $l \gg \lambda$. We have therefore undertaken certain additional measurements of the line shape under conditions (2). The results constitute the content of the first part of this paper.

The different mechanisms for electron scattering can be assumed to be independent in the first approximation, and l can be represented in the form

$$1/l = 1/l_0 + 1/l_T(T), \tag{3}$$

where l_0 is the mean free path at 0°K and is caused by the static violations of the lattice periodicity, while l_T is the free path determined by the electron-phonon collisions. As is well known, in such collisions the electron is scattered through a small angle $\beta \approx p_{ph}/p_F$ (p_{ph} is the phonon momentum). On the other hand, each type of SE has its own angle $\theta^{(i)}$ of "maximum possible scattering," and when this angle is exceeded the electron ceases to contribute to the amplitude of SE line. Inasmuch as β is comparable with $\theta^{(i)}$, the efficiency of the electron-phonon collisions may turn out to be different for different types of SE. Then the measured numerical values of l_T will also be different. Moreover, even for a concrete type of SE, the value of $\theta^{(i)}$ depends on the ratio δ/d (δ—depth of skin layer), and sometimes also on the magnitude of the magnetic field H[2], which may affect the measurements of l/T.

Since interest attaches not to the functions $l_t(T)$ in each concrete case, but to the path $l_{e,\,ph}^{(T)}$ traversed by the electron between two collisions with phonons, we

have attempted to ascertain under what conditions l_T can be reliably identified with $l_{e,ph}$. For a comparison of the numerical results, we measured the statistical electric resistivity $\rho(T)$ on the same samples as used for the observation of the SE, using the method of rotating magnetic fields[10,11].

EXPERIMENTS

Although the SE manifests itself in the same manner in the real and imaginary parts of the surface impedance of plates $Z = R + iX$, it is more convenient to use for amplitude measurements the quantity X. The sensitivity of apparatus to the measurement of R is determined by the amplitude generated by the radial oscillator in whose tank circuit the investigated sample is connected, and requires a constant correction when the temperature or the magnetic field is varied. To the contrary, the changes of the generation frequency $\Delta f \sim -\Delta X$ do not depend on the generation regime, and no calibration is required for relative measurements of the amplitude of the SE lines.

The apparatus employed is described in[12]. Modulation coils were mounted independently of the electromagnet, so that the modulating field did not rotate relative to the surface of the sample when the inclination of the magnetic field was varied. Since the smallest tilt of the coils led to the appearance of a signal $\partial f/\partial H$ that increased monotonically with increasing field H, the modulating field was positioned parallel to the surface of the sample to within a fraction of a degree by minimizing the signal in the strong field. When desired, the tilt of the coils could be used to compensate for the monotonic part of the change of the impedance in the vicinity of the SE line.

The potassium was obtained from KNO_3 reduced with iron with subsequent vacuum distillation. The distillation was in a glass sphere of approximately 12 mm diameter, which was sealed off and placed on a torsion balance with a rotating magnetic field to measure the resistance[10,11]. The field ranged from 5 to 15 Oe, the rotary frequency at room temperature was of the order of 100 Hz, and at helium temperatures on the order of 0.02–0.002 Hz. The measurement accuracy at low temperatures was 0.5%.

The samples were made either by pouring liquid metal in a dismountable glass mold (samples 1 and 3) or by compressing a piece of potassium between two glass plates. In either case, the glass was coated with a layer of mineral oil to prevent the metal from sticking to the glass when solidified. The prepared samples were washed in gasoline, and then the gasoline was evacuated, and the sample was cooled in vacuum to the temperature of liquid nitrogen, at which it was customarily stored. In the instrument, the samples were inserted in the apparatus likewise at liquid-nitrogen temperature. After the high-frequency measurements were completed, samples 1 and 3, which had the form of regular discs of 18 mm diameter, were again mounted on a torsion balance. This made it possible not only to verify that the potassium was not contaminated during the course of preparation of the samples, but also to measure directly the value of $\rho(T)$ of the sample with which the SE was investigated. The characteristics of the samples are listed in the table. For the purest samples $\rho_{room}/\rho_0 \approx 6 \times 10^3$ (ρ_0—residual resistance).

Line Shape

The results of the study of the line shape can be summarized as follows. At relatively small free path ($l < d, \lambda$), the SE line shape can vary with the ratio between the parameters δ, l, and d.

The dependence on l can be demonstrated most easily in Fig. 1 for the SE line in a field H inclined to the surface of the metal at an angle $\varphi = 25°$. As shown in[9] and[14], at an inclination angle $\varphi \approx 25°$, the SE line from a spherical Fermi surface is formed as a result of the focusing of a large group of electrons moving along different trajectories and arriving from one side of the plate to the other along paths of different lengths, so that $\Delta\lambda/\lambda \approx 0.4$ (see Fig. 2; $\lambda_{max} = 2d/\sin 2\varphi$). At 4°K, the SE line is formed mainly as a result of the shorter trajectories of type b. Lowering of the temperature causes the electrons to start to arrive at the opposite side of the plate also along longer trajectories a, as a result of which the line shape changes.

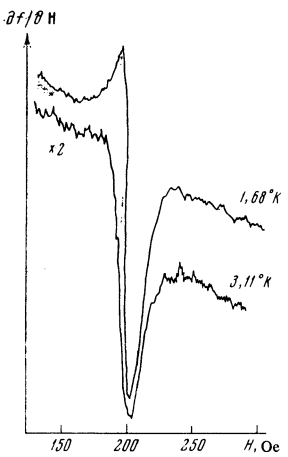

FIG. 1. Plot of SE lines for sample 1, $\varphi = 25°$. Modulation field parallel to the surface of the metal. Projection of H on the plane of the sample is parallel to the high-frequency current j. Frequency f = 4.1 MHz. The lower curve is plotted with the sensitivity increased by a factor of 2.

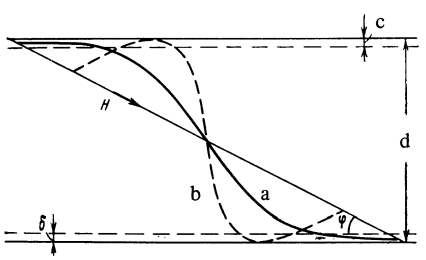

FIG. 2. Curve a—boundary orbit separating the effective orbits from the ineffective ones [14], b—orbit with two effective points per period.

On the Fermi sphere, this is the only case of focusing of trajectories of different length, although for an arbitrary Fermi surface such a focusing should probably be encountered quite frequently. All the remaining SE lines observed by us for potassium did not

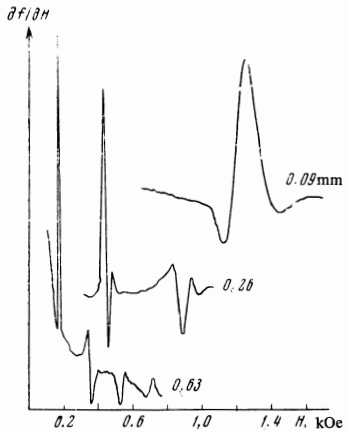

FIG. 3. Plots of SE lines of samples 1, 4, and 5, $\varphi = 0$, $\mathbf{j} \perp \mathbf{H}$, $f \approx 7$ MHz, $T = 1.3°$ K. The ordinate scales are different.

Sample number	d, mm	$l_0^{(\rho)}$, mm	l_0, mm	$10^2\ \delta/l$	$10^2\ \delta/d$
1	0.63	0.19		3.2—2.6	1.0—0.8
2	0.23	0.13		6.5	3.7
3	0.24	0.13		4.1	2.2
4	0.26	0.13	0.12	3.8	1.9
5	0.09	0.13	0.16	3.6	5.2
6	0.25	0.06		8.5	2.0

Note. The depth of penetration δ was determined from the Se line width: $\delta \approx \approx 0.15\, d\Delta H/H$ [3,13].

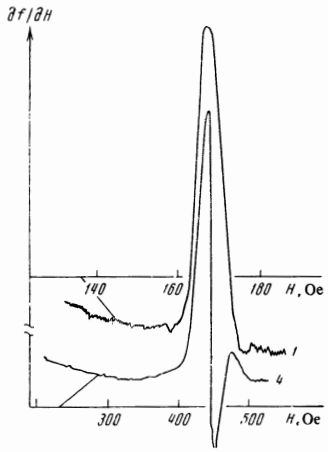

FIG. 4. Comparison of Se lines of samples 1 and 4, $\varphi = 0$, $\mathbf{j} \perp \mathbf{H}$, $f \approx 6$ MHz, $T = 1.9°$ K. The ordinate scales are different.

change their shape with temperature. However, in measurements of $l(T)$ this circumstance must always be checked, for other mechanisms governing the dependence of the line shape on T are quite probable. For example, for sufficiently broad lines $\Delta H/H_0 \approx 10\%$ (H_0—field corresponding to the left edge of the line), an important role may be played by the fact that in the right part of the line, in a field $H = H_0 + \Delta H$, the effective trajectory connects one of the surfaces of the plate not to the other surface, but to the internal part of the skin layer located near it, so that the role of d is played by the quantity $d - \delta \approx d(1 - \Delta H/H_0)$. Because of this, the right-hand and left-hand extrema of the line could, generally speaking, vary with the temperature in different manners. In our experiments we did not observe such an effect, probably because the broad lines were obtained by us at not too small values of d, when condition (2) was violated.

There exists also other causes for changes of the line shape. As seen from Fig. 3, the SE line shape is different for samples of different thickness. These changes are not connected with the multiplicity of the return of the electron to the skin layer and are inherent apparently in SE of all types. This indicates, in particular, that for all the samples inclination of the field by ~15° did not change the shapes of the lines shown in the figure, and only shifted them towards stronger fields[8]. It should be particularly noted that in two samples, 4 and 6, the purity of which differs by a factor of 2, the SE lines obtained for equal δ and d were identical in shape, although their amplitudes differed by one order of magnitude.

Of course, the difference in l between samples 4 and 6 is not so appreciable as to influence noticeably the distribution of the electromagnetic field in the skin layer. On the other hand, however, it is not likely that this distribution can be influenced also by the interaction between the skin layers under conditions when $\delta/d \approx (1 - 2) \times 10^{-2}$ (see the table). It follows therefore that when $l < d$ the SE line shape does not reflect the distribution of the field in the skin layer, as is the case in the opposite limiting case $l \gg d$ (compare with[13]), and depends on some complicated relations between the parameters δ, d, and l. The meaning of this conclusion is seen particularly clearly from Fig. 4, where plots of two SE lines in samples of different thickness (samples 1 and 4) are drawn in abscissa scales proportional to d^{-2} (we recall that the line width is $\Delta H \sim H_0\delta/d \sim \delta/d^2$).

Measurements of Free Path

a) Results. The measurements of l_0, obtained by determining the dependence of the amplitude of SE lines at the limiting point on the angle φ of inclination of the magnetic field to the surface of the sample, were performed on sample 4 in the angle interval $14.5° < \varphi < 20°$ and on sample 5 in interval $8.5° < \varphi < 14.5°$. The measurements were made at $T = 1.3°$K, when the phonon part of the scattering is no longer significant. Reduction by means of the formula

$$A = A_0 \exp(-\lambda/l_0) = A_0 \exp(-2d/l_0 \sin 2\varphi)$$

led to values of l_0 that coincided practically with the values of $l_0^{(\rho)}$ obtained from measurements of the static conductivity (see the table; $l_0^{(\rho)}$ was calculated from the residual resistance ρ_0 by means of the formula $l^{(\rho)} = p_F/e^2 N \rho_0$, and l_0 was obtained from SE experiments). This agreement means that the SE and the dc conductivity are equally sensitive to scattering by static inhomogeneities.

Typical results of the measurement of l_T by the SE method are shown in Fig. 5 in coordinates $2(\ln A)/\pi d = \text{const} - 1/l_T$ and T^3. It is seen from the figure that all the samples except the thinnest sample 5 lead to a relation $l_T = bT^{-3}$, whereas for the first SE line on the central section (trajectory diameter $D = d$) there were obtained on samples 1—4 values of b from 2.7 to 3.0 cm-deg^3.

The lower two straight lines of Fig. 5 pertain to SE lines on the central section in double and triple fields

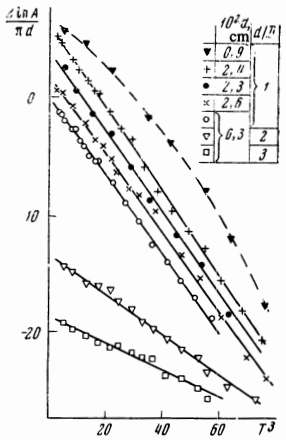

FIG. 5. Temperature dependence of the amplitude of the SE lines, measured in a parallel field. The zero on the ordinate axis pertains to the straight line drawn through all the symbols O. The random measurement error can be assessed from the scatter of the points.

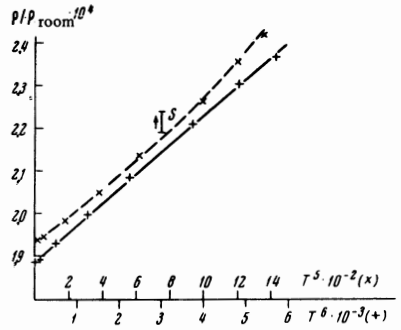

FIG. 6. Electric resistivity ρ as the function of T^5 (X) and T^6 (+). The $\rho(T^5)$ curve is shifted upward along the ordinate axis by a distance S.

(D = d/2 and d/3). When reduced by means of formula (1) with $\alpha = \pi/2$, they yielded for b the values 1.5 and 1.0 cm-deg^3, i.e., smaller by factors 2 and 3 respectively than the values measured on the first line. From the temperature dependence of the amplitude of the SE line on a section close to the central one, at an inclination angle $\varphi = 17°$, the value obtained for sample 4 was b = 1.7 cm-deg^3.

In sample 5 the relation is in the form $l_T \sim T^{-4}$, not only in the central section but also at the limiting point.

The results of the measurements of the resistance $\rho(T)$ of a spherical sample of potassium sealed in glass are shown in Fig. 6. Since the shape of the sample necessary to obtain the absolute values of the resistance by this method must be regular, we confine ourselves to relative measurements. The absolute values of ρ were obtained by starting from the value $\rho_{room} = 7.19 \times 10^{-6}$ Ω-cm (see[15]). As seen from Fig. 6, in the interval 1.3–4.2°K the resistivity of potassium varies like $\rho(T) \sim T^6$. The T^6 dependence was observed already in the measurements of $\rho(T)$ of pure sodium[16]. The fact that the T^5 dependence has been observed so far for potassium[16,17] is apparently due to the insufficient purity of the investigated material.

b) Discussion. The cubic dependence in $l_T(T)$ is usually evidence that a single collision with a phonon makes an electron ineffective. At the same time, the customarily employed estimate for the angle of the "maximum permissible scattering" $\theta^{(c)}$ on the central section, namely $\theta^{(c)} \sim \sqrt{\delta/d}$, leads to the inequality $\theta^{(c)} > \beta$, which should result in the relation $l_T \sim T^{-5}$, owing to the diffuse character of the scattering.

Let us examine, however, in greater detail the different possibilities of scattering of an electron on the central section by a phonon. Figures 7a and b show three essentially different cases: the electron goes over to the neighboring orbit in momentum space without changing the phase of rotation (process 1), it shifts along the previous orbit remaining in the vicinity of the point $v_z = 0$ (process 2; the z axis is normal to the surface of the sample) or in the vicinity of the point $v_z = v_{z.max}$ (process 3). The effectiveness of the process 1 is determined by the change of the diameter of the trajectory $D = d - \delta = d(1 - \theta_1^2/2)$, whence $\theta_1 \sim \sqrt{\delta/d}$. The process 2, the effectiveness of which is

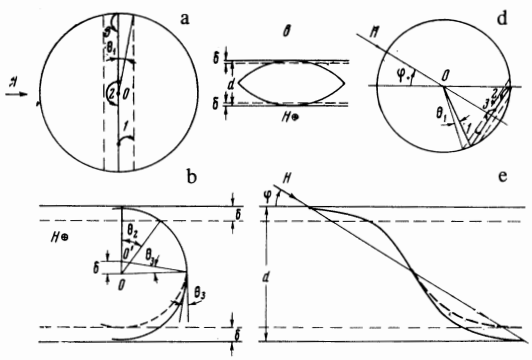

FIG. 7. Drawings a and d pertain to momentum space, while b, c, and e pertain to coordinate space. The dashed lines designate the skin layers and the trajectories of the electrons after scattering.

connected with the change of the time of stay in the skin layer, i.e., with the width of the effectiveness strip (Fig. 7b), leads to a similar estimate $\theta_2 \approx \theta_1$. On the other hand, the effectiveness of process 3 is determined by the shift of the center of the trajectory along the direction of the electron velocity at the instant of scattering, and it can be readily seen from Fig. 7b that $\theta_3 \approx \delta/d$.

It follows from the foregoing that in the vicinity of the point $v_z = v_{z.max} \approx |v|$ there exists on the central trajectory a region of most effective scattering. In our experiments we have the relation

$$\delta/d \lesssim p_{ph}/p_F < \sqrt{\delta/d}. \qquad (4)$$

Therefore the observed $l_T \sim T^{-3}$ dependence is due apparently only to scattering processes of the third type, whereas the remaining collisions with the phonons are not very effective. For the thinnest sample (sample 5), the left side of the inequality (4) is violated and the scattering acquires a diffuse character already on the entire trajectory, and consequently the degree of T increases.

It is curious that Naberezhnykh and Tsymbal[5], who obtained $l_T \sim T^{-5}$ for cadmium, dealt with a "lenslike" trajectory with a kink, in which $v_z \ll |v|$ throughout (Fig. 7c), and consequently scattering processes of the third type are not very effective.

All the foregoing is applicable also to the SE at the turning point. The estimate $\theta^{(0)} \sim (\delta n/d)^{2/3}\varphi$ (n = 1,

2, 3, ... is the number of the line), given in[2], starts from the width of the effectiveness band and is valid only for processes 1 and 2 (Fig. 7b), whereas for processes 3 (Fig. 7e), which shift the axis of the trajectory, we have $\theta_3^{(0)} \sim (\delta n/d)\varphi$. The difference here, however, is somewhat smaller than on the central section, and furthermore all the estimates of $\theta^{(0)}$ contain an additional small factor φ, which made it possible to obtain in[1,2] effectiveness of all the collisions.

It might seem that in the approximation (2) the amplitudes of the SE lines on the central section in multiple fields should depend on l in the same manner as the first line. Indeed, when $D = d/n$ ($n = 1, 2, 3, \ldots$) the field amplitude in the burst closest to the surface is $q \sim \exp(-\pi d/2n)$, and the amplitude of the SE line is $A \sim q^n$[18]. However, for sample 1, in the temperature interval 3.9—1.3°K, the first SE line in a parallel field increased by six times, the second by 2.6 times, and the third by only 1.9 times. The same values of l_T for different n are obtained by reducing the experimental data in accordance with formula (1) with $\alpha = \pi/2n$, but in spite of the attractiveness of such a reduction method, there seems to be no justification for it. Inasmuch as for the first number at different values of d the results agree with each other, the discrepancy for different values of n cannot be ascribed to the change of the parameter δ/D with changing number. It is possible that the measurements made at $n \neq 1$ cannot be reduced in principle by means of formula (1), since the probability that the electron will execute one revolution without being scattered when $D = d$ is equal to the probability of executing n revolutions at $D = d/n$, and the amplitudes of SE lines of all numbers are of the same order.

In view of the lack of a reliable method of reducing the amplitudes of the lines with higher numbers, we confine ourselves to a consideration of the values of b obtained with lines with $n = 1$, namely $b = 2.8$ cm-deg^3. Inasmuch as we obtained a T^6 dependence for the electric resistance, which is unexpected from the theoretical point of view, let us compare first the results obtained for b with the estimates of the electron-phonon range $l^{(\kappa)}$, obtained from data on the electronic heat conductivity κ[19] and the specific heat C[20]: $l_{e,ph} \sim l^{(\kappa)} \approx 3\kappa/Cv \approx 0.35\ T^{-3}$ cm (the Fermi velocity is $v = 0.74 \times 10^8$ cm/sec). Starting from the fact that the value of l_T observed by us in the SE is due to collisions of type 3, amounting to approximately 1/4 of the total number of collisions, we obtain $l_{e,ph} \approx l_T/4 \approx 0.7^{-3}$ cm. The discrepancy between our results in parallel and inclined fields is apparently due to the change of the fraction of the effective collisions. Taking into account the approximate character of all the numerical estimates, the agreement between our results and $l^{(\kappa)}$ should be regarded as perfectly satisfactory. Thus, we have succeeded in measuring in potassium the average free path of the electron between two collisions with phonons with the aid of the SE on the central cross section. We hope that in the future the availability of purer potassium will enable us to verify once more the results with the aid of the SE at the limiting point.

We now turn to the temperature dependence of the static resistance. The only mechanism to us, leading to a stronger temperature dependence of the electric resistance than T^5, is connected with the quenching of the U-processes of scattering in the electron-phonon system[21]. It is not at all surprising that at temperatures lower than 40°K the U-processes have not yet been fully quenched out, since, in view of the fact that the minimum distance from the Fermi Sphere to the boundary of the Brillouin zone is $P = 0.173\ \hbar\pi/a$ (a—lattice constant), and the velocity of the slow transverse wave along the corresponding direction (the [110] axis) is $s = 0.65 \times 10^5$ cm/sec[22], we need a phonon with energy $2Ps \sim 10°$ for the Unklapp process. However, if we turn to the numerical values of the free path, additional difficulties arise. If the phonon system is in equilibrium, the quantity $l_T^{(\rho)}$ entering in the formula

$$\rho(T) - \rho(0) = p_F/Ne^2 l_T^{(\rho)},$$

should be equal to

$$l_T^{(\rho)} \approx l_{e,ph}(\Theta/T)^2 \approx 4 \cdot 10^3 T^{-5}\,\text{cm}$$

($\Theta \approx 90°$ is the Debye temperature). If the $\rho \sim T^6$ dependence is due to the quenching of the U-processes, this means that there is dragging by the phonon system. But in this case the effectiveness of the electron-phonon collisions should decrease, so that $l_T^{(\rho)}$ should become still larger at all temperatures. Our experimental result is of the form

$$l_T^{(\rho)} = 3.4 \cdot 10^2 T^{-6}\,\text{cm},$$

so that even at 1°K the value of $l_T^{(\rho)}$ is one-tenth the value of $l_{e,ph}(\Theta/T)^2$, instead of being larger. Similar discrepancies between $l^{(\kappa)}$ and $l_T^{(\rho)}$ in alkali metals were already pointed out by Klemens[23]. Discrepancies of the same sign were observed also for other metals[2,3].

The authors are grateful to Academician P. L. Kapitza for the opportunity of working at the Institute of Physics Problems, to Professor Yu. V. Sharvin for help in organizing the work and for valuable advice and discussions, to G. M. Kolosova for purifying the KNO_3 salt, and to I. F. Poletaev for consultations on the procedure of obtaining potassium.

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Translated by J. G. Adashko
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