VARIATION OF THE ELECTRICAL RESISTANCE OF SOLID MERCURY AT LOW TEMPERATURES

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The temperature dependence of the electrical resistance of solid mercury was investigated in the temperature ranges 1.6—5.1°K and 14—23°K. It is shown that, in the helium range of temperatures, the electrical resistance R could be described by the equation $R = R_0 + AT^n$; when the temperature was lowered from 5.1 to 1.6°K and the purity increased by a factor of almost 100, n increased and became equal to 5 for pure samples at $T \leq 3$°K. Very pure mercury exhibited unique properties not shared by other metals: in the temperature range from 4.2 to 1.7°K, its resistance changed by a factor of 50. This strong temperature dependence of the resistance began below 20°K. The influence of a longitudinal magnetic field (up to 2.5 kOe) on the electrical resistance of mercury was determined for the first time.

THE temperature dependence of the electrical resistance is known in considerable detail for very many metals. The exception is mercury, which has been mentioned only in Andrew's paper,[1] where it is reported briefly that the resistivity of mercury in the helium range of temperatures (4.2—1°K) is proportional to 4.2d power of the temperature. There is also an earlier paper by Sckell[2] on the resistance of mercury single crystals in the temperature range from −45.5°C to −191.5°C. Sckell determined the value of the electrical resistivity ρ of mercury single crystals along two principal crystallographic directions; the orientation was also determined from the resistivity. To determine the orientation, Sckell[2] measured the resistivity of 210 samples of randomly differing orientations, from which he determined the values of the maximum and minimum resistivity. Moreover, he grew several single crystals in the form of three mutually perpendicular cylindrical wires and determined the average value of the resistivity sums for each trio of such wires, which should be constant and equal to B. Using the results obtained and the fact that $\rho_x + \rho_y + \rho_z = \rho_{\parallel} + 2\rho_{\perp} = B$, Sckell found the values of $\rho_{\parallel}$ (along a ternary axis) and $\rho_{\perp}$ (at right-angles to the ternary axis) for all the temperatures that he investigated.

In these two investigations, the samples of solid mercury were kept all the time in glass capillary tubes. The dearth of investigations of the electrical conductivity of mercury is due to the difficulties of preparing mercury single crystals, of determining their orientation, and in handling them.

The present paper describes an attempt to extend our knowledge of the electrical conductivity of solid mercury at low temperatures. The electrical resistance of mercury was investigated mainly at liquid helium and hydrogen temperatures using mercury samples of different degrees of purity.

SAMPLES AND EXPERIMENTAL TECHNIQUE

Mercury samples in the shape of cylindrical wires of about 2.5 mm diameter and 80—100 mm long were grown from mercury of the R-2 grade and 99.99% purity (in accordance with the State Standard GOST 4658-49).

The majority of the samples listed in Table I were grown by the Stepanov method[3] and only four samples (Nos. 10, 14—16) were prepared by the Bridgman method in glass tubes, which were washed first in ethyl alcohol.[4] In the latter case, a grown sample was extracted from a glass tube with the aid of an alcohol "lubricant" by heating the sample and the tube to a temperature of ≈ 100°K. Samples Nos. 1, 3—7 were grown using a single crystal of very pure bismuth as a seed. Samples Nos. 9, 11, 13 were grown using a mercury single crystal, prepared by pulling a bismuth seed from the melt. Sample No. 12 was grown using a pure single-crystal seed. Samples Nos. 2 and 8 were obtained by determining of samples Nos. 4 and 16, respectively.

Table I. Principal electrical properties of all samples.

Sample No.	$\frac{R_0}{R_{77}} \cdot 10^3$	$\frac{R_{4.2}}{R_{77}} \cdot 10^3$ *	$\frac{R_{4,2}}{R_{1,7}}$	$\frac{R_{4.2}}{R_0}$	$\frac{R_{5,1}}{R_{1,6}}$ **	$\frac{R_{5,1}}{R_0}$ **	n	T ***
1	1.67	7.82	4.1	5.2	—	—	3.5	4.2
2	1.54	8.05	4.4	5.2	—	—	3.5	4.2
3	1.12	7.27	5.2	6.5	9.7	12.5	3.5	4.2
4	1.06	7.35	5.2	7.0	9.0	13.5	3.2	5
5	0.71	7.22	7.0	10.2	11.3	18.4	3.5	4
6	0.41	6.42	7.6	14.8	—	—	3.5	4.2
7	0.198	6.00	15.4	30.0	29.6	5.7	4	4
8	0.16	6.15	19.2	38.0	—	—	3.7	4.2
9	0.111	6.18	24.0	55.7	—	—	4.5	3.5
10	0.096	5.83	28.5	60.7	62	115	4.5	3
11	0.087	5.68	28.0	66.0	68.5	148	4.5	4.2
12	0.074	5.73	31.5	76.5	71.5	148	4.7	3
12a	0.035	5.73	43.0	160.0	103	310	4.7	3
13	0.0625	5.72	33	91	—	—	4.2	4.2
14	0.0605	5.82	26	96.5	—	—	4.2	4.2
15	0.034	5.90	31	174.0	—	330	4.2	3.5
16	0.0218	5.81	51.5	267.0	125	507	5	3

*The average value is $R_{4.2}/R_{77} = 5.85 \times 10^{-3}$, if the averaging is carried out over all samples after subtracting first the value of the corresponding residual resistance. This value can be regarded as the ideal relative resistance of mercury at 4.2°K.

**Between 4.2 and 5.1°K, the resistance of mercury increases by a factor of 1.9. This value is the average of the measured values for nine samples after subtracting R_0/R_{77} from $R_{4.2}/R_{77}$ and $R_{5.1}/R_{77}$ for the least pure samples.

***T is the highest temperature at which the indicated power exponent applies.

The samples grown were usually single crystals, as indicated by the slip bands which appeared in the samples after accidental bending, by the Laue diffraction patterns recorded for some samples, and by the high plasticity, not observed for polycrystalline samples. The crystallographic orientation of the samples was not known.

To select the best method for the preparation of the samples and their electrical mounting, it was necessary to estimate the influence of accidental mechanical bending on the electrical resistance of mercury. For this purpose, several samples of mercury of various purities were bent eight times through an angle of ≈90° so that they assumed a zigzag shape but the degree of deformation was not known. Measurements of the resistance of these samples in the helium range of temperatures before and immediately after deformation showed that the influence of the deformation was greater for the purer samples and at lower temperatures. The relative change in the resistance due to deformation was 5—15% at 4.2°K and 60—70% at 1.8°K. This showed that, even though accidental deformation would have been less than that produced in the samples by deliberate bending, mercury samples had to be handled carefully so as not to deform them during mounting. For the same reason, the Bridgman method of growing should not be used because a sample is deformed at the moment of extraction from the glass tube.

To measure the electrical resistance, we attached the samples to bakelite sheets so that they could freely distend or contract without deformation during heating or cooling. The resistance was measured by a compensation method using copper for the current contacts (0.3 mm diameter), which were fused into the sample, and for the potential contacts (0.1 mm diameter), which were welded to the sample by a spark discharge.

The resistance was determined using an R-306 potentiometer and a M 17/3 galvanometer of ≈5 $\times 10^{-8}$ V voltage sensitivity. The measuring current was 60 mA at room temperature and 6 A at helium temperature. Our studies showed that variation of the measuring current from 3 to 10 A at 4.2 and 2°K had no influence, within the limits of the experimental accuracy, on the resistance of the purest samples. The error in the measurement of the resistance of the purest samples did not exceed 0.5% at 4.2°K and 6% at 2°K.

The temperature intervals 1.6—5.1 and 14—23°K were reached by pumping away the vapor in the helium and hydrogen baths, respectively, and by increasing the pressure in the baths up to 1.3 excess atmospheres in a metal cryostat. The temperature corresponding to a selected vapor pressure was determined from the 1958 tables[5] for helium and hydrogen. To obtain temperatures of 77 and 196.6°K (−73.5°C), we used liquid nitrogen and solid carbon dioxide. To measure the latter temperatures, we used a copper resistance thermometer whose resistance was 15.42 Ω at 20°C. Oscillations of the nitrogen temperature did not exceed 0.2—0.4°K.

The magnetic field for the destruction of the superconductivity of the mercury samples was produced by a solenoid whose constant was 52.5 Oe/A. In the middle part of the solenoid (20 cm long), where all the measurements were carried out, the field was 2.5 kOe and its uniformity was within 1%.

RESULTS

The influence of the purity of mercury[1)] (represented by different values of the relative resistance R_0/R_{77} of the metal) on the power exponent of T and on the magnitude of the fall of the resistance of mercury in the temperature range from 4.2 to 1.7 and 0°K is demonstrated in Table I and in Fig. 1. Instead of the usual ratio R_T/R_{293}, we used everywhere the ratio R_T/R_{77}, where R_T is the resistance of a sample at a selected temperature.

The reason for the use of the ratio R_T/R_{77} instead of R_T/R_{293} was this. According to Sckell's data,[2] at the melting point the resistance of mercury increases on the average by a factor of 4.35 and the increase is different for samples of different orientations (the differences are up to 14% from the average). We did not know the crystallographic orientation of the samples and, consequently, we did not know the conversion coefficient at the melting point. Therefore, our constant "high temperature" was the boiling point of nitrogen, which was sufficiently high to suppress the influence of impurities and defects on the value of the resistance of mercury.[2)] The relative resistance at higher temperatures, for example at 234°K (−39°C), could be determined by a simple calculation from the known value of $R_{234}/R_{77} = 3.37$.[3)] Thus, it was found that for the purest samples $R_0/R_{234} = 6.5 \times 10^{-6}$ and for the least pure samples $R_0/R_{234} = 5 \times 10^{-4}$.

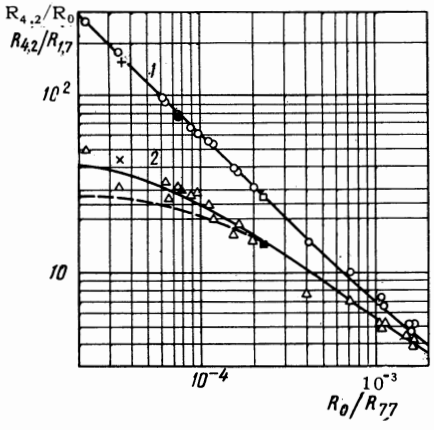

FIG. 1. Dependence of the relative fall of the resistance in the temperature range from 4.2 to 1.7 and 0°K on the residual resistance R_0/R_{77}. Curves 1 and 2 show, respectively, the variation of the quantities $R_{4.2}/R_0$ and $R_{4.2}/R_{1.7}$. The dashed curve gives values of $R_{4.2}/R_{1.7}$ calculated for $R_{1.7}(T)/R_{77} = 1.9 \times 10^{-4}$. ●▲ – Sample No. 12 (in a magnetic field); +× – sample No. 12a (same sample No. 12 without magnetic field), □■ – Andrew's results.[1]

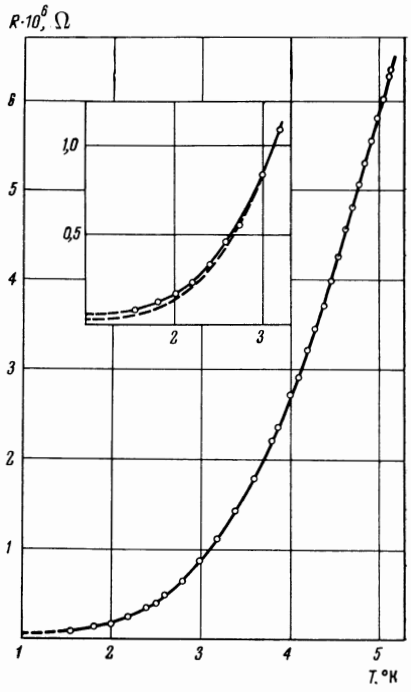

FIG. 2. Dependence of the resistance R on temperature for sample No. 12. The dashed curve shows the dependence of R on T on an enlarged (resistance) scale after subtracting the influence of a longitudinal magnetic field.

The results given in Fig. 1 and Table I were not corrected for the influence of the external magnetic field. This correction would only have increased the value of the change in the resistance in the interval from 4.2 to 1.6 and 0°K.

The temperature dependences of the resistance of mercury samples are presented in Figs. 2 and 3. The power exponent n of T, the residual resistance R_0, and the coefficient A were determined by selecting the values for these quantities so as to obtain the best agreement between the points calculated from the empirical equation $R = R_0 + AT^n$ and the experimental curve. The error in such a deter-

1)The mercury became contaminated by the partial solution of the bismuth seed in the melt when the mercury crystal was pulled.

2)This is also true of other easily melted metals such as Sn, In, Pb, Cd, Zn.[6]

3)This value was obtained by extrapolation using a linear law for the points R_{77}, R_{196}, R_{228}.[2]

mination of n was ±0.05. For the purest sample (No. 16), the resistance was $R \propto T^{3.4}$ in the temperature range 5.1—4°K; $R \propto T^{4.2}$ in the range 4—3°K; and $R \propto T^{5}$ in the range 3—1.6°K. For the least pure sample (No. 1), we found $R \propto T^{3.5}$ over the whole temperature range 1.6—4.2°K. The dependence of the exponent on T can be seen clearly from the dependences of R on T plotted in logarithmic coordinates after subtracting the residual resistance. For this reason, Fig. 3 shows the dependences for the purest and least pure samples.

We estimated the influence of a longitudinal magnetic field on the resistance of mercury samples in the helium temperature range. For this,

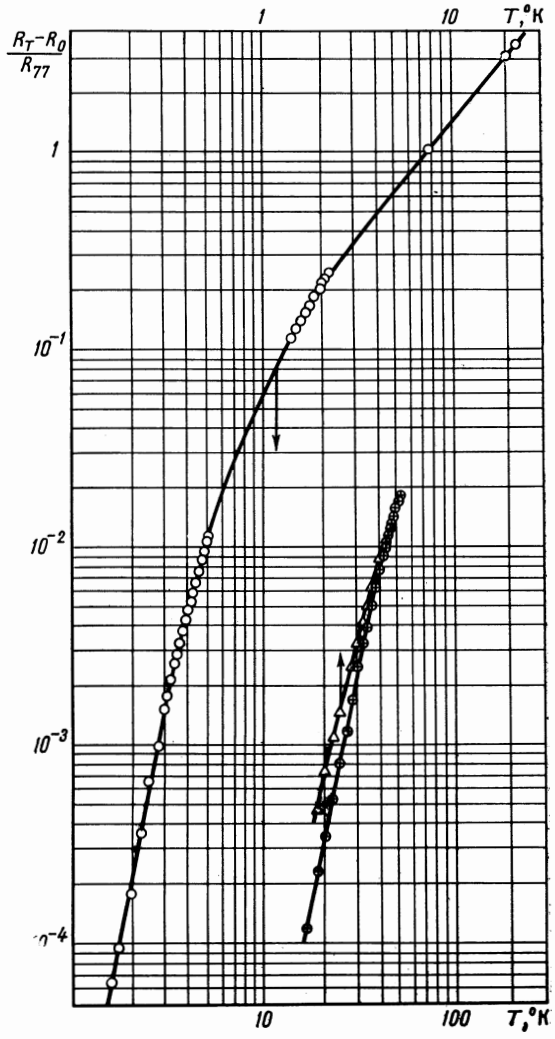

FIG. 3. Temperature dependence of the ideal electrical resistance of mercury samples of three different purities. To avoid overlap of the curves, they were shifted horizontally by one order of magnitude with respect to one another. The vertical arrows point to the appropriate temperature scale. ○ – sample No. 16, $R_0/R_{77} = 2.2 \times 10^{-5}$ (in a magnetic field); Δ – sample No. 12, $R_0/R_{77} = 1.67 \times 10^{-3}$ (in a magnetic field); ⊕ – sample No. 12a, $R_0/R_{77} = 3.5 \times 10^{-5}$ (without a field).

we used a fairly pure sample No. 12 with $R_0/R_{77} = 7.4 \times 10^{-5}$ (in a magnetic field) and recorded the dependence of R on the magnetic field between H = 2.5 kOe and the critical field H_{cr} at T = 4.2, 3, 2.75, 2.5, 2.25, 2.0, 1.8, and 1.6°K. The maximum value of H_{cr} at the lowest temperature in our experiments was only ≈360 Oe,[7] but nevertheless it was necessary to estimate its influence on the temperature dependence of the resistance. The R(H) curves obtained at a given temperature were extrapolated from H_{cr}, using a quadratic law, to zero magnetic field.

The extrapolation of all the curves showed that, for sample No. 12 with $R_0/R_{77} = 7.4 \times 10^{-5}$, the increase in the resistance due to the external magnetic field was 12.5% at 1.8°K and 1.5% at 3°K. In the case of samples that were three times less pure, such as the samples used by Andrew,[1] this correction was very small (≈4%) even at 1.8°K. However, in the case of the purest samples, for example No. 16, the increase in the resistance at 1.8°K could reach 30—40%. Unfortunately, the dependence of the resistance on the field could not be determined in detail for this sample.

After correction for the influence of the longitudinal magnetic field, the curve showing the dependence of R on T for sample No. 12 changed only at T < 3°K, as shown in Fig. 2. The residual resistance decreased by a factor of 2 (cf. Table I, samples Nos. 12 and 12a) while $R_{4.2}/R_0$ and $R_{4.2}/R_{1.7}$ increased by a factor of $1\frac{1}{2}$—2 (cf. Fig. 1) but there was practically no change in the power exponent n of T.

Using the curves R(H) and the extrapolated values $R_{H=0}$ for each temperature, we plotted for the first time ever the dependences $\Delta R/R_{H=0}$ on H (Fig. 4), where ΔR is the change in the resistance

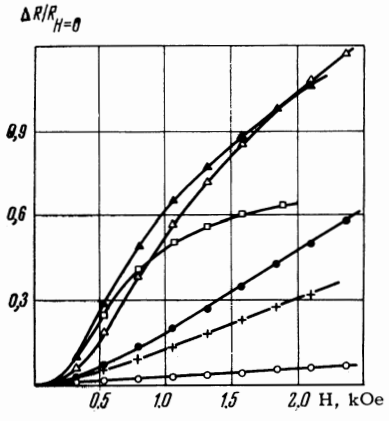

FIG. 4. Relative change in the resistance of sample No. 12 as a function of a longitudinal magnetic field and temperature. ○ – 4.2°K, + – 3°K, ● – 2.75°K, Δ – 2.5°K, ▲ – 2°K, □ – 1.8°K.

Table II. Change in resistance between 4.2 and 0°K for various metals.

	Hg	Ga[8]	Tl	Pb[9]	Sn[10]	Cs[11]	In[9]	Cd[9]	Sb	Bi	Zn[9]	Rb[11]
$R_{4,2}/R_0$	260	16	5	3,3	3,3	2,4	2.15	2	40%	30%	25%	23%
R_0/R_{293}	$6.5\cdot10^{-6}$*	$5,4\cdot10^{-7}$	$2,3\cdot10^{-5}$	$1,9\cdot10^{-5}$	$3\cdot10^{-6}$	$2\cdot10^{-3}$	$3.4\cdot10^{-5}$	$1\cdot10^{-5}$	$4.5\cdot10^{-4}$	$3.5\cdot10^{-3}$	$1,1\cdot10^{-5}$	$2.6\cdot10^{-3}$
t_{mp}, °C	—39	29	303	327	232	28,5	156	321	630	271	419	39
Θ, °K	75	125	88	88	111	50	106	134	142	80	214	69

*The value of the ratio R_0/R_{234}.

due to the application of a parallel magnetic field.

DISCUSSION OF RESULTS

The most interesting property of mercury is the very large drop of the resistance in the relatively narrow temperature range ($\Delta T = 3.5°K$) between 5.1 and 1.6°K. This drop in the resistance increases with the purity of mercury and reaches two orders of magnitude for the purest samples (cf. Table I). Figure 1 shows the change in the resistance in the temperature range from 4.2 to 1.6 and 0°K. The very large increase in this drop is clearly evident from Fig. 1: with decreasing R_0/R_{77} the resistance changed by a factor of 40—50 in the range from 4.2°K to 1.6—1.7°K, and the change from $R_{4.2}$ to R_0 (the latter was a calculated value) was by a factor of 170—270 (cf. Table I).

Curve 1 in Fig. 1 is described by the equation

$$\frac{R_{4,2}}{R_0} = 1 + \frac{R_{4.2}(T)/R_{77}}{R_0/R_{77}},$$

which is, in fact, the well known Matthiesen rule: $R_{4.2} = R_0 + R_{4.2}(T)$, where R_0 is the residual resistance, $R_{4.2}(T)$ is the lattice resistance at 4.2°K, and $R_{4.2}$ is the total resistance at 4.2°K. In our calculations, we assumed $R_{4.2}(T)/R_{77} = \rho_{4.2}(T)/\rho_{77} = 5.85 \times 10^{-3}$ (cf. Table I). Curve 2 should be described by the equation

$$\frac{R_{4,2}}{R_{1.7}} = \left[1 + \frac{R_{4,2}(T)/R_{77}}{R_0/R_{77}}\right] \Big/ \left[1 + \frac{R_{1,7}(T)/R_{77}}{R_0/R_{77}}\right],$$

but to calculate it we had to know the value of $R_{1.7}(T)/R_{77} = \rho_{1.7}(T)/\rho_{77}$, which was not known with sufficient accuracy. Comparison of the calculated and experimental curves indicated that the best agreement was obtained for $R_{1.7}(T)/R_{77} = 1.9 \times 10^{-4}$, although a considerable discrepancy was still observed for the pure samples.

In other words, the curves presented in Fig. 1 show the dependence of the change in the resistance in the temperature range from 4.2 to 0°K on the purity of the metal. These curves can be used to estimate the purity of mercury (from the resistance), for which $R_{4.2} \approx R_{1.7} \approx R_0$. The purity of such mercury corresponds to $R_0/R_{77} \approx 10^{-2}$ or $R_0/R_{234} \approx 3 \times 10^{-3}$.

From the results obtained, we could deduce that, of all the known metals, mercury exhibits the largest change in the resistance in the temperature range 4.2—0°K. This is evident from Table II, which lists all the metals whose resistance changes by more than 20% when the temperature is lowered from 4.2 to 0°K. All the metals listed in Table 2 have low melting points and low Debye temperatures Θ. The data presented in Table II and the results reported in[6] suggest that the values of $R_{4.2}/R_0$ should rise with increasing purity of all these metals, particularly those of Cs and Rb.

It is evident from Fig. 3 that the resistance of mercury varies linearly with temperature in the range from 234 to 30—40°K. A similar observation, referring to the temperature range 240—77°K, has been reported earlier.[2] Because of the very low Debye temperature, the liquid hydrogen range of temperatures is still very high in the case of mercury and, therefore, the resistance falls only by a factor of 17 between 234 and 20.4°K ($\Delta T = 215°K$). Only below 20°K does the resistance of mercury begin to vary strongly (between 20.4 and 5°K, the resistance decreases by a factor of 20). Finally, a very rapid fall of the resistance begins in the helium range of temperatures (from 5.1 to 0°K), where the resistance decreases by a factor of ≈500 (cf. Table 1). It is interesting to note that the resistance of mercury at 4.2°K is not a function of its purity if the sample is not too contaminated. To judge the purity, it is better to use the resistance at ≈1.5°K, although even at this temperature the resistance of mercury is still 2—4 times greater than the residual value.

It is evident from Table I that the power exponent of T goes up with the increasing purity of mercury and the region where a given R(T) law is obeyed becomes narrower and shifts in the direction of lower temperatures. If we compare the results obtained with those of Andrew,[1] whose mercury samples had $R_0/R_{77} = 2.1 \times 10^{-4}$, we find that his conclusion of $R \propto T^{4.2}$ in the helium range of temperatures is in good agreement with the re-

sults for sample No. 7 (cf. Table 1).

Since the Debye temperature of mercury is low, the range of temperatures in which the resistance changes from $R \propto T$ to $R \propto T^5$ extends right down to 3°K, while for the majority of metals which can be melted fairly easily but have a higher Debye temperature (Sn, In, Pb, Tl, Cd), the lower limit of thsi interval lies above 4.2°K.[9] For this reason, the resistance of pure mercury in the helium temperature range cannot be described by a single law either in the form of the empirical equation $R = R_0 + AT^n$ or in the form of the well-known theoretical relationship $R = R_0 + \alpha T^2 + \beta T^5$. Only in the case of very pure mercury at $T \leq 3$°K do we observe the Bloch dependence $R \propto T^5$. The resistance due to the scattering of electrons on other electrons ($R \propto T^2$) should evidently appear at temperatures $T \leq 1.6$°K.

Some idea about the degree of influence of a longitudinal magnetic field up to 2.5 kOe on the resistance of mercury at various temperatures can be obtained from Fig. 4. The curves in that figure show that when the temperature is reduced to 2.5°K the relative change in the resistance in the magnetic field increases at first but then it decreases. The increase in $\Delta R/R$ with the reduction of the temperature is associated with an increase in the average mean free path of conduction electrons (as the effective magnetic field is increased) and the decrease at $T < 2.5$°K can evidently be explained by the effect of the dimensions of a sample—the size effect (because of the very large increase in the mean free path when temperature is lowered). A similar effect has been observed already for Sn, Zn, Al[12] and In.[13] Bearing in mind that the diameter of mercury samples was 2.5 mm, it follows that the mercury is quite pure and has a mean free path comparable with the diameter of a sample. This conclusion requires a careful experimental check.

We attempted to plot Kohler's diagram. However, it was found that the curves obtained differed in their slope and the experimental points for different samples did not fit a single curve. This was evidently due to the strong anisotropy of the electrical resistance in a longitudinal magnetic field, which was first demonstrated for Sn, Cd, and Zn in[12].

The reported semi-quantitative estimates of the influence of the mechanical deformation on the electrical resistance show that the Matthiesen rule is not satisfied in the temperature range 4.2—1.6°K, namely the quantity

$$\Delta\delta = (R_T/R_{77})_{\text{deform}} - (R_T/R_{77})_{\text{undeform}}$$

decreases by a factor of 1.6—1.8 when the temperature is reduced. We may expect that the influence of deformation on the power exponent n of T is similar to the influence of impurities, i.e., this exponent should decrease. However, no reliable data have yet been obtained on this point.

Finally, the increase in the value of the resistance at 77°K with the increase in the number of measurements (number of cooling runs from 230 to 77°K), reported by Sckell,[2] was not observed even in the helium range of temperatures, which indicated that the measurements must be carried out on mercury samples without any shielding containers.

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