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TEMPERATURE DEPENDENCE OF THE RESISTANCE OF FILAMENTARY ZINC CRYSTALS

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The temperature dependence of the resistance $\rho(T)$ of thick and thin single crystals of zinc (whisker thickness up to 0.6 μ) is investigated between 1.5 and 300°K. No significant difference in the law of increase $\rho = \rho(T)$ is detected in bulk ($d \gg \lambda_{4.2}^{(\infty)}$) and thin ($d \ll \lambda_{4.2}^{(\infty)}$) samples. The residual resistance is attained in whiskers below 4°K with an accuracy of up to 1 percent. Between 5 and 12°K the behavior of the resistance can be described by the law $\rho = \alpha T^{4.4 \pm 0.2}$ for whiskers and $\rho \propto T^{4.6 \pm 0.2}$ for bulk samples. Between 12 and 20°K $\rho = \beta T^{3.2 \pm 0.2}$ for whiskers and $\rho \propto T^{3.8 \pm 0.2}$ for bulk samples. A transition region exists up to approximately 80°K; above this temperature $\rho \propto T$ in both cases. The resistance temperature coefficients α and β of whiskers depend on the sample thickness. They are inversely proportional to the thickness of filamentary whiskers. The dependence is less pronounced in plates. The experimental results are not in accord with the theoretical predictions. In the present paper this discrepancy is explained by the fact that 1) electrons moving at small angles to the surface are reflected by the latter almost specularly and as a result of this the Olsen mechanism is absent; 2) the mean specular reflection coefficient for all the electrons depends on the temperature and tends to zero with increasing temperature.

THE effect of the transverse dimensions d of a metal sample on its electrical resistance can to a first approximation be written in the following form:[1]

$$\rho^{(d)} = \rho^{(\infty)} + \rho^{(\infty)}\lambda^{(\infty)}d^{-1}f(p), \qquad (1)$$

where the superscripts d and ∞ refer to thin (of thickness d) and bulk ($d \gg \lambda^{(\infty)}$ is the electron mean free path in the bulk sample, p is the coefficient of specular reflection of electrons from the surface ($1 \geq p \geq 0$), f(p) is a function of p whose form depends on the relationship between $\lambda^{(\infty)}$ and d.

For completely diffuse reflection (p = 0) the function f(p) = 1; for completely specular reflection (p = 1) the function f(p) = 0. For $\lambda^{(\infty)} \gg d$ one usually neglects the term $\rho^{(\infty)}$ in expression (1). Since this term will be important for $p \approx 1$, we shall retain it for the sake of generality of the discussion. The product $\rho^{(\infty)}\lambda^{(\infty)}$ is a constant quantity that does not depend on the temperature. Therefore, in accordance with (1) (with an accuracy to within a few percent) the temperature dependence of the resistance in thin samples should not depend on the thickness and the slope of the straight line $\rho^{(d)} \propto d^{-1}$ should not depend on the temperature. Measurements carried out by a number of authors[2–4] on thin samples of indium, mercury, cadmium, tin, and aluminum have shown that the slope of the straight line $\rho^{(d)} \propto d^{-1}$ changes with temperature more appreciably than should be expected from calculations in accordance with [1]. Thus in aluminum[4] it increases by 70 percent for a temperature increase from 4 to 20°K.

Expression (1) has been obtained under the assumption that the scattering cross section of electrons on phonons does not depend on the angle between the direction of motion of the electron and the surface of the sample. However, this condition cannot be fulfilled at low temperatures, a fact first pointed out by Olsen.[2] According to Olsen, when $1 \gg T/\Theta \gg d/\lambda^{(\infty)}$ (Θ is the Debye temperature) the collision of an electron traveling at an acute angle to the surface with a phonon leads to its collision with the surface. As a result the momentum of the electron changes by a quantity comparable with the momentum itself. The Olsen mechanism should increase the efficiency of electron-phonon collisions in a thin sample at low temperatures and should lead to a considerable increase of the thermal part of its resistance compared with a bulk sample. Quantitative calculations employing this mechanism were carried out by Blatt and Satz[5] as well as by Azbel' and Gurzhi.[6] Below we shall dwell on the conclusions of [6] since these results were obtained in the most general case—both for wires and plates.

In a thin sample ($d \ll \lambda^{(\infty)}$) at low temperatures the effective collision frequency of the electrons drifting along the surface with phonons is greater than in a bulk sample $\nu_{ef}^{(d)} \gg \nu_{ef}^{(\infty)}$. In a bulk sample $\nu_{ef}^{(\infty)} \propto (T/\Theta)^3 \times (T/\Theta)^2$ where the factor $(T/\Theta)^3$ takes into account the number of phonons and $(T/\Theta)^2$ is due to the small collision efficiency of the electron and the phonon (the scattering at a small angle is of the order of T/Θ). In a thin sample for glancing electrons (the angle between the direction of motion and the surface is smaller than $d/\lambda^{(\infty)}$) the collision with a low-temperature phonon is efficient since it leads to a collision with the surface where diffuse scattering is assumed. Therefore the factor $(T/\Theta)^2$ cancels out for glancing electrons and $\nu_{ef}^{(d)} \propto (T/\Theta)^3$. In view of the fact that in thin samples the current is chiefly transferred by glancing electrons, the electron-phonon mean free path $\lambda_{ef}^{(d)} \propto (\Theta/T)^3$, and correspondingly the thermal part of the resistance $\rho^{(d)}(T) \propto (T/\Theta)^3$. This should be valid under the condition

$$T/\Theta \gg d/\lambda_{ei},\ (d/\lambda_{ef}^{(\infty)})^{1/3}, \qquad (2)$$

where λ_{ei} is the mean free path due to impurities.

For any relationship between $d/\lambda^{(\infty)}$ and T/Θ the resistance of a thin sample with completely diffuse reflection is given by the following expressions: for platelets

$$\rho^{(d)} \approx \rho^{(\infty)} + \frac{\rho^{(\infty)}\lambda^{(\infty)}}{d}\left[\ln\left(1+\frac{1}{x_0}\right)\right]^{-1}, \qquad (3)$$

for wires

$$\rho^{(d)} \approx \rho^{(\infty)} + \frac{\rho^{(\infty)}\lambda^{(\infty)}}{d}(1+\delta); \qquad (4)$$

here

$$\delta = \frac{d}{\lambda_{ei}}\ln\frac{1}{x_0} + \frac{1}{2}\frac{d}{\lambda_{ef}^{(\infty)}}\left(\frac{\Theta}{T}\right)^2 \ln\left[1+\left(\frac{T}{\Theta}\right)^2\frac{1}{x_0^2}\right]$$

$$x_0 = d\left(\frac{1}{\lambda_{ei}} + \frac{1}{\lambda_{ef}^{(\infty)}}\right) + \frac{d}{\lambda_{ef}^{(\infty)}}\left[\left(\frac{T}{\Theta}\right)^2 + \left(\frac{d}{\lambda_{ef}^{(\infty)}}\right)^{2/3}\right]^{-1}.$$

An analysis of these formulas shows that in the general case the residual resistance in thin samples is attained at considerably lower temperatures than in bulk samples. In the region of very low temperatures ($T/\Theta < d/\lambda_{ei}$), $\rho^{(d)}(T) \propto T^5$ and under condition (2) we have $\rho^{(d)}(T) \propto T^3$. At higher temperatures the resistance increases according to a logarithmic law and then again becomes proportional to T^5, as in a bulk sample.

This work was undertaken for the purpose of observing the above laws in whiskers (filamentary crystals) of zinc whose thickness amounted to several microns, and with a mean free electron path at 4.2°K of $\lambda_{4.2}^{\infty} \geq 300\,\mu$. Such measurements on whiskers have been carried out for the first time.

METHOD OF MEASUREMENT

The technique of preparing and mounting the whiskers was described by us in [7, 8]. For the measurements we chose both filamentary whiskers (FW) as well as whiskers in the form of strips (SW). Some of the characteristics of the measured samples are given in the Table.

In order to obtain temperatures from 300 to 4.2°K use was made of the natural temperature gradient inside the liquid-helium dewar. For this purpose a disc-like thick-walled copper chamber was attached to the lower end of a long rod made of Monel tubing passing through the dewar cover; inside the chamber was a holder with the whisker or bulk sample of zinc. The desired temperature could be obtained by moving the chamber above the level of liquid helium. The temperature was checked with the aid of a gold with iron and chromel thermocouple. The thermocouple was calibrated with a gas thermometer. Its thermal emf in the range of 4–300°K is close to linear; the sensitivity is about 17 μV/deg. The thermocouple was attached to the holder of the whisker or directly to the bulk sample. As the chamber was slowly lowered the emf of the thermocouple and of the sample was recorded by a two-coordinate chart recorder. Below 30°K, in order to retain the required accuracy, the measurements were carried out point by point with potentiometer circuits.

MEASUREMENT RESULTS

In the range from room temperature to $T \approx 80$°K the resistance of bulk samples and whiskers varies within

No. of sample and orientation	d*, μ	$\lambda_{4.2}^{(\infty)}$** $/d$	$\rho_{4.2}^{(d)}/\rho_{295}^{(d)} \cdot 10^3$	n in the law $\Delta\rho_T \propto T^n$	
				20–12° K	12–5° K
Bulk samples					
1, Polycrystalline	1500	0.04	0.5	3.8	—
2, [11$\bar{2}$0]	1000	0.25	0.11	3.5	4.5
3, [11$\bar{2}$0]	1200	0.25	0.1	3.8	4.6
4, [0001]	1000	0.5	0.05	4.0	4.6
Filamentary whiskers***; $\lambda_{4.2}^{(\infty)} \geqslant 300\,\mu$, $\rho_{4.2} \geqslant 0.5$; the axes are parallel to the (1$\bar{1}$00) plane and produce with the [0001] axis angles of 60° (preferentially) and 40° ([11$\bar{2}$1] and [11$\bar{2}$2]).					
1	1	300	8	3.1	4.5
2	1.3	230	5.8	3.0	4.3
3 ****	1.4	214	11	3.3	—
4	1.4	214	6.5	3.0	4.4
5 ****	1.75	170	8.5	3.3	4.4
6	2.1	143	3.7	3.2	4.5
7	2.4	125	3.2	3.2	4.4
8	2.5	120	3.0	3.1	—
9 ****	2.6	115	6.9	3.0	4.3
10	3.95	76	2.0	3.3	4.6
11	3.95	76	2.1	3.0	4.6
Plate-like whiskers***; axes parallel to the (0001) plane					
12	0.58	518	3	3.3	4.3
13	0.78	385	1.6	—	—
14 ****	1.3	230	1.7	3.2	4.3
15	1.6	188	1.2	3.4	4.6

* For FW $d = \sqrt{s}$ where s is the cross-sectional area of the whisker determined from the resistance at 295°K and the length of the sample. For SW $d = s/b$ where b is the width of the platelet. The accuracy of the determination of d is ± 10 percent.

** The reduced values $\leqslant \lambda_{4.2}^{(\infty)}/d$.

*** The information about $\lambda_{4.2}^{(\infty)}$ and $\rho_{4.2}$ and the orientation of the whiskers was obtained in [8].

**** The whiskers had defects introduced during the mounting.

an accuracy of 4 percent in proportion to the temperature: $\rho(T) = A + BT$. Below 80°K down to temperatures of 20–25°K a transition region is observed: the $\rho(T)$ dependence changes from a linear to a cubic law $\rho \propto T^3$. For the temperature range from 20 to 4°K the results are shown in Fig. 1 in which we have plotted on the ordinate axis on a logarithmic scale the difference of the resistivities $\Delta\rho_T^{(d)} = \rho^{(d)}(T) - \rho^{(d)}(0)$ where $\rho^{(d)}(0)$ is the residual resistivity of the samples; the value of $\rho^{(d)} \times (0)$ was found by extrapolation of the results to 0°K in such a way that for $T < 10$°K the largest number of experimental points fell on a single straight line $\Delta\rho_T^{(d)} \propto T^n$.

As the processing of the experimental $\Delta\rho_T^{(d)}(T)$, curves showed, in the investigated range of temperatures the behavior of the resistances of bulk and thin zinc samples obeys approximately the same power laws (see the Table). In the 12–20°K temperature range one can write $\Delta\rho_T^{(d)} = \beta T^{3.2 \pm 0.2}$ for whiskers and $\Delta\rho_T^{(\infty)} \propto T^{3.8 \pm 0.2}$ for bulk samples; in the 5–12°K range $\Delta\rho_T^{(d)} = \alpha T^{4.4 \pm 0.2}$ and $\Delta\rho_T^{(\infty)} \propto T^{4.6 \pm 0.2}$ respectively. The values of the exponents were averaged over all the samples. The indicated accuracy in their determination is due mainly to this.

For whiskers the temperature coefficients α and β depend on the thickness of the samples. In FW the dependence is observed very clearly,[1)] in SW it is weaker.

[1)] The effect of different crystallographic orientations encountered in FW [8] on the coefficients α and β is not considered since in accordance with the measurements of Aleksandrov [4] it can be expected that it does not exceed the experimental errors.

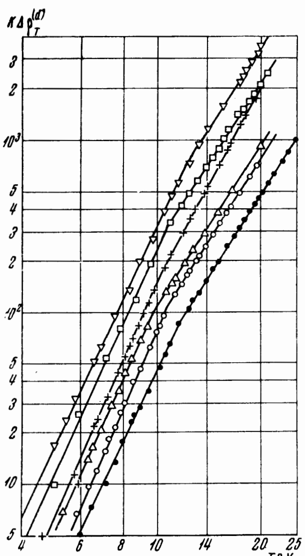

FIG. 1

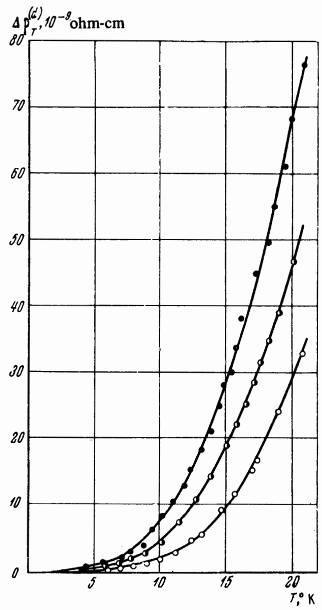

FIG. 2

FIG. 1. Temperature dependence of the resistance for whiskers and for a bulk sample. Constant multipliers K (given in units of 10^{10} ohm^{-1} $-cm^{-1}$) have been introduced for convenience in presenting the results. Samples: ∇ – SW No. 12, K = 7.3; Δ – FW No. 3, K = 1.8; □ – FW No. 1, K = 3.2; ○ – FW No. 10, K = 1.9; + – bulk sample No. 3, K = 8.1; ● – FW No. 7, K = 1.2.

FIG. 2. The thermal part of the resistivity of zinc samples: ● – FW No. 1, ◑ – FW No. 7, ○ – bulk sample No. 3.

In order to illustrate this, we present in Fig. 2 the temperature dependence of $\Delta\rho_T^{(d)}$ for different thicknesses of the samples, and in Fig. 3 the dependence of $\Delta\rho_T^{(d)}$ on two fixed temperatures. It is seen in Fig. 3 that for FW one can write

$$\Delta\rho_T^{(d)} = \Delta\rho_T^{(\infty)} + \gamma d^{-1}, \qquad (5)$$

where the coefficient γ depends on the temperature, increasing with it. It was found that within the limits of accuracy of the measurements expression (5) is valid at any temperature within the 6–20°K range.

The accuracy in measuring $\rho^{(d)}$ for whiskers (0.1–0.01 percent for different samples) made it possible to observe the variation of the resistance in the region below 4.2°K. Unfortunately it turned out that these changes are related not only with the properties of the investigated whiskers but mainly with the effect of the voltage contacts which introduced a nonreproducible additional resistance of the order of 1 percent which made it impossible to carry out measurements at $T < 4°K$.

DISCUSSION OF THE RESULTS

Of the investigated temperature ranges the one of most interest is the range below 12°K. In discussing the results obtained in this range we shall assume that the thermal part of the resistance is only related to the normal processes of the scattering of electrons by phonons. We shall then write a Bloch–Grüneisen type of law in the region below 12°K in the form

$$\Delta\rho_T^{(\infty)} \propto (T/\Theta)^{4.6}. \qquad (6)$$

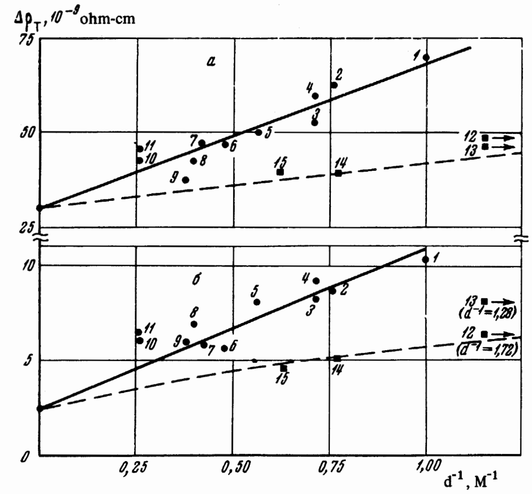

FIG. 3. Dependence of the thermal part of the resistivity on the reciprocal of the sample thickness at two temperatures: a – T = 20°k, b – 11°K. Solid lines (points ●) – FW, dashed lines (■ squares) – SW.

We shall determine from condition (2) and from the dependence (6) the maximum thickness and the minimum temperature for which one should according to formulas (3) and (4) observe below 12°K the dependence $\Delta\rho_T^{(d)} \propto T^3$. Let us choose the following values: the Debye temperature $\Theta = 240°K$, $\lambda_{ei} \approx \lambda_{4.2}^{(\infty)} = 300\ \mu$,[8] $T_{max} = 12°K$, and $\lambda_\Theta^{(\infty)} = 4 \times 10^{-6}$ cm [obtained from a measurement of $\rho^{(\infty)}$ at T = 240°K and from the value of $\rho^{(\infty)}\lambda^{(\infty)} = 1.8 \times 10^{-4}$ ohm-cm].[4,8] Then the condition $T_{max}/\Theta \geq (d/\lambda_{ef}^{(\infty)})^{1/3}\,\lambda_{ef}^{(\infty)} = \lambda_\Theta^{(\infty)}(\Theta/T)^{4.6}$, leads to the value $d_{max} \leq 5\ \mu$, and from the condition $T_{min}/\Theta \geq d_{max}/\lambda_{ei}$ we find that $T_{min} \geq 4°K$.

One could thus hope that in whiskers whose thickness is less than 5 μ and bulk samples in the 4–12°K range one will observe a considerable difference in the law governing the temperature variation of the resistance. However, this conclusion has not been confirmed by the experimental results. The $\Delta\rho^{(d)}(T)$ dependence in bulk and in the very thinnest samples in the 5–12°K range follows approximately the same power law $\Delta\rho_T \propto T^{4.5}$. The same result was obtained by Wyder[9] for indium in an investigation of the temperature variation of the resistance of thin films and bulk samples: in both samples: in both instances $\Delta\rho_T \propto T^5$.

According to the conclusions of the work of Azbel' and Gurzhi[6] one could also have expected that the ratio $\Delta\rho_T^{(d)}/\Delta\rho_T^{\infty}$ below 12°K will be of the order of magnitude of $(\Theta/T)^{1.6}$. For instance, at 11°K this ratio should be approximately 150. It is seen from Fig. 3 that even for the thinnest measured FW $\Delta\rho_{11°K}^{(d)}/\Delta\rho_{11°K}^{(\infty)} \approx 4$.

Let us now consider the dependence of $\Delta\rho_T^{(d)}$ on the the thickness of the sample. For FW under condition (2) $\delta \approx (d/\lambda_T^{(d)})\ln(\lambda_T^{(d)}/d)$ where $1/\lambda_T^{(d)} = 1/\lambda_{ei} + 1/\lambda_T^{(\infty)} \times (T/\Theta)^2$. Furthermore, we find from (4) that

$$\Delta\rho_T^{(d)} \approx \Delta\rho_T^{(\infty)} + \rho_T^{(\infty)}\ln(\lambda_T^{(d)}/d) \propto \ln(d^{-1}).$$

The experimental results shown in Fig. 3 indicate rather that the dependence of $\Delta\rho_T^{(d)}$ on d^{-1} is close to linear.

We assume that the reason for the indicated inconsistencies between the experimental results and the conclusions of [6] is the fact that the Olsen mechanism is not operative in zinc whiskers. This can only be explained by the nature of the reflection of electrons by the surface of the metal. In all the work devoted to this problem it is assumed that the reflection from the boundaries is completely diffuse. This also refers to "glancing" electrons. It is precisely the diffuse nature of the reflection which should ensure under the appropriate conditions that the factor of the type $(T/\Theta)^2$ cancels out in the expression for the resistance $\Delta\rho_T^{(d)}$. Clearly, for completely specular reflection (p = 1) the thermal parts of the resistance of bulk and thin samples should coincide, $\Delta\rho_T^{(\infty)} = \Delta\rho_T^{(d)}$. As shown by measurements, in zinc whiskers at T = 4.2°K even the average coefficient of specular reflection $p \geq 0.5$. One can therefore assume that at low temperatures electrons moving at acute angles to the surface undergo almost completely specular reflection. The temperature dependence of the resistance of thin samples cannot in such a case be changed by "glancing" electrons.

How is it to be explained that a difference in the temperature variation of the thermal part of the resistance of bulk and thin samples is nevertheless observed? This can only be related with the fact that the magnitude of the coefficient p is temperature dependent. As is known, the parameter p describes the averaged properties of the sample surface.[10] It is assumed that one p-th of the electrons is specularly and the remainder diffusely reflected. The value of p depends, roughly speaking, on the "roughness" of the surface, or, in other words, on the degree of treatment of the latter. However, the "roughness" of a surface should also depend on the temperature. In fact at high temperatures, when the phonon and electron momenta are of comparable magnitude, even reflection of electrons by the most perfect surface should be diffuse. Bearing this in mind for the low-temperature region, the coefficient p should be presented in the form of two parts—temperature dependent and independent:

$$p = p_0 - p_T = p_0(1 - p_T / p_0).$$

In analogy with the resistance, p_T should depend on the number of phonons and the efficiency with which they scatter electrons. One can therefore write $p_T \propto (T/T_{dif})^n$ where T_{dif} defines the temperature for which reflection becomes completely diffuse. The power exponent should of course depend on which phonons scatter the electrons at the surface of the sample. If these are volume phonons then the exponent should be the same as in the law $\Delta\rho^{(\infty)} \propto (T/\Theta)^n$. Assuming that this is true for zinc whiskers, one can estimate T_{dif} making use for this purpose of experimental values. Such an estimate was carried out in accordance with the following formula:[1]

$$\rho_T^{(d)} = \rho_T^{(\infty)} + \frac{\rho_T^{(\infty)}\lambda_T^{(\infty)}}{d}\left(\frac{1-p}{1+p}\right). \quad (7)$$

It led to a value of $T_{dif} < 100°K$.

It must be noted that expression (7) explains well the behavior of resistance with temperature in filamentary whiskers: the power law, the decrease in the slope of the the straight lines $\rho^{(d)} \propto d^{-1}$, and the dependence of $\Delta\rho_T^{(d)}$ on the thickness.

According to this point of view, the thermal parts of the resistance of bulk and thin samples should coincide for completely diffuse reflection (p = 0 in the entire temperature range), whereas the maximum possible difference between them should be observed for completely specular reflection (at very low temperatures).

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