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Resistivity of Thin Metallic Wires*)

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Abstract. Measurements by OLSEN on thin indium wires have demonstrated that not only the residual but also the temperature dependent part of the resistivity increases with decreasing wire diameter. It was suggested by OLSEN that small angle electron-phonon scattering may account for the observed effect by scattering electrons to the surface where they suffer diffuse reflection.

Since an exact solution of the transport equation, taking account of such phonon-surface scattering events, is beset with mathematical difficulties we have considered the problem using basically the same elementary approach as employed by Nordheim. We find that Olsen's mechanism leads to an additional resistivity given by

$$\varrho_{ps} = (2\pi)^{1/3} (mv_F/ne^2)^{2/3} (T/\theta_D)^{2/3} [\varrho_i^N(T)]^{1/3} (r)^{-2/3};\ r \ll l.$$

Here v_F is the velocity at the Fermi energy and $\varrho_i^N(T)$ is the ideal resistivity due to Normal phonon scattering only. This expression is valid only if r, the wire radius, is less than the electron mean free path, l, in the bulk material.

The above expression is in reasonable agreement, qualitatively and quantitatively, with Olsen's results and, within the range of validity, also accounts for Andrew's observations. It is suggested that careful measurements on thin wires may allow an experimental separation at low temperatures between the ideal resistivities due o Normal and to Umklapp processes.

I. Introduction

The resistivities of thin films and wires are known to differ appreciably from the bulk resistivities whenever the physical dimensions of the specimen become comparable to or smaller than the electronic mean free path (m. f. p.) in the bulk. The additional resistivity arises from scattering of electrons at the external surfaces of the sample, and theory has shown that careful measurements of the resistivity of wires as a function of diameter should allow a direct determination of the mean free path and may also shed light on the nature of the scattering process at the surface.

The earliest theories[1)2)] of the size effect were based on rather elementary m. f. p. concepts. Subsequently, FUCHS[3)] and DINGLE[4)] con-

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sidered the resistivities of thin metallic films and of wires of circular cross section in a more satisfactory way. In their theories the surfaces play the role of boundary conditions imposed on the solution of the Boltzmann equation.

The expression for the resistivity of a thin wire derived by DINGLE cannot be reduced to analytic form in the general case. However, numerical evaluation shows that this 'exact' result differs from Nordheim's approximate expression

$$\varrho_W = \varrho_b + \alpha(\varrho_b\, l/d) \tag{1}$$

by no more than 5% over the entire range $0 < l/d \leqslant \infty$. Here ϱ_W is the resistivity of the wire, ϱ_b the resistivity of the substance in bulk, l is the bulk m. f. p. and d is the diameter of the wire. The factor α in Equation (1) depends on the nature of the scattering process at the surface. If reflection at the surface is specular $\alpha = 0$, if it is diffuse $\alpha = 1$. Experimental results indicate diffuse reflection for most metals[5][6]). (BISMUTH[7]) plays its perennial role of exception to the rule.) We shall set $\alpha = 1$ in all subsequent expressions.

The striking prediction of Equation (1) is that, to the conduction electrons in a thin wire, the external surfaces play the same role as impurities, resulting in a temperature independent residual resistivity

$$\varrho_s = \varrho_b\, l/d. \tag{2}$$

We recall here that

$$A \equiv \varrho_b l = mv_F/ne^2 \tag{3}$$

is practically temperature independent. Thus we may write for the resistivity of a thin wire

$$\varrho_W = \varrho_i(T) + \varrho_0 + \varrho_s \tag{4}$$

where $\varrho_i(T)$ is the ideal, ϱ_0 the residual resistivity of the bulk material.

According to Equations (2) and (4) Matthiessen's rule applies, and consequently the *temperature dependent* part of the resistivity of a thin wire should be the same as that of a bulk sample. Recent measurements by OLSEN[8]) have shown that such is not the case. In Figure 1 we display Olsen's results so as to focus attention on the failure of Matthiessen's rule. We have plotted there the difference $\varrho_W(T) - \varrho_W(0)$. If Equation (4) were valid all the curves should overlap. Instead, the apparent ideal resistivity of the thinner indium wires is considerably greater than that of those of larger diameter, the difference exceeding by far the 5% error attributable to the use of the approximate relation, Equation (1). A similar discrepancy between theory and experiment, noted by ANDREW[9]), was dismissed

by DINGLE[4]) who attributed it to differences in the method of preparation of the very thin and somewhat thicker mercury wires. Some of Andrew's results are shown in Figure 2, where, again, we are plotting the difference $\varrho_W(T) - \varrho_W(0)$. In view of OLSEN's work, Andrew's data on the thinner wires deserve reexamination.

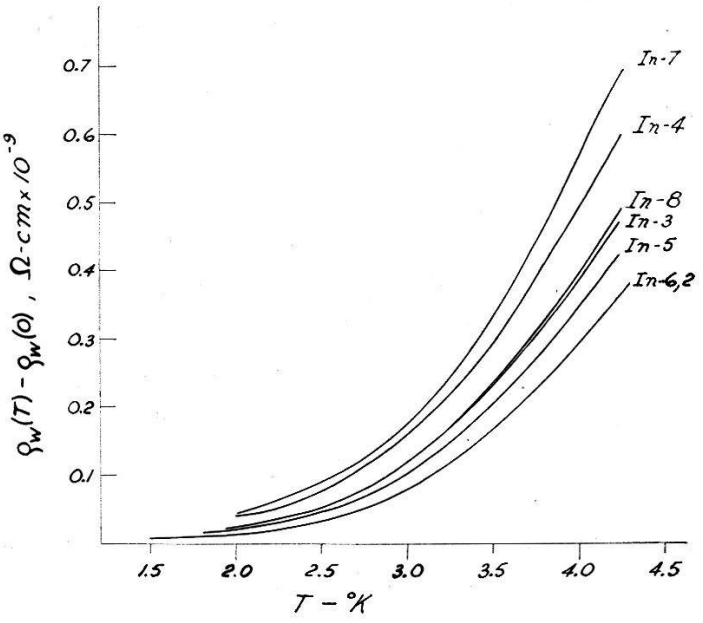

Fig. 1

The temperature dependent part of the resistivity of indium wires, as determined by OLSEN. The sample specifications are given as

Specimen	Source	diameter, mm
In – 2	JOHNSON-MATTHEY	2·0
In – 3	Tadanac Brand	0·311
In – 4	Tadanac Brand	0·086
In – 5	Tadanac Brand	0·57
In – 6	Tadanac Brand	2·54
In – 7	JOHNSON-MATTHEY	0·06
In – 8	JOHNSON-MATTHEY	0·20

OLSEN suggested the following mechanism which might account for the enhanced temperature dependence of the resistivity of thin wires. In the bulk, electron-phonon scattering of the Normal type (non-Umklapp) can scatter electrons at low temperatures only through rather small angles θ, where

$$\theta \leqslant \theta_{\max} \simeq (T/\theta_D)\,(K_B/k_F).$$

Here K_B is the radius of the Brillouin Zone in the spherical approximation (i. e. K_B is the wave vector of an acoustical phonon of energy $k\theta_D$) and k_F is the wave vector of an electron at the Fermi surface. For most

metals (omitting from consideration here Bismuth and similarly misbehaved semi-metals) $(K_B/k_F) \sim 1$, and we then have

$$\theta \leqslant \theta_{max} \simeq T/\theta_D \tag{5}$$

At low temperatures randomizing of linear momentum by normal electron-phonon scattering requires a large number of electron-phonon collisions, and consequently the effective m. f. p. is rather long. In a thin wire the electrons which, in a given small interval of time, are primarily responsible for the flow of charge are those whose momentum is very nearly parallel to the wire axis. All other electrons must shortly reach the surface and there suffer diffuse reflection. Electrons moving along the wire axis in a thin wire are, however, far more susceptible to small angle phonon scattering than are electrons in the bulk. A few small angle events may deflect such an electron sufficiently to bring it to the surface where, though on impact it is still moving largely in the 'forward direction', it suffers diffuse reflection and loses all memory of its past history. Thus, the presence of the surface greatly enhances the effectiveness of small angle phonon scattering, particularly as it limits the m.f.p. of those electrons which, at any instant of time, are the important charge carriers.

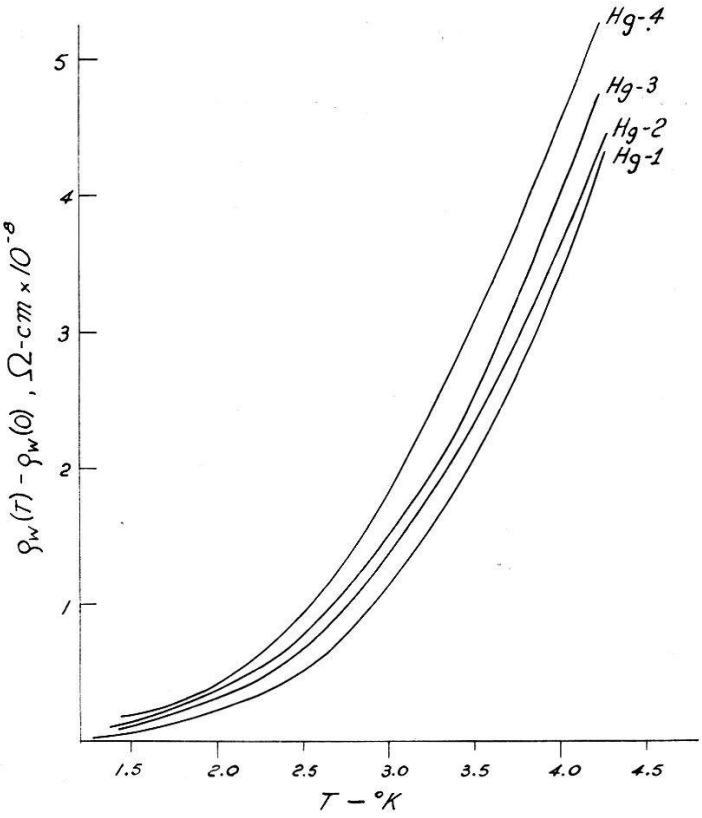

Fig. 2

The temperature dependent part of the resistivity of mercury wires, as determined by ANDREW. The wire diameters are Hg-1, d = 0·79 mm; Hg-2, d = 0·0166 mm; Hg-3, d = 0·0105 mm; Hg-4, d = 0·0061 mm.

In the following we shall demonstrate that this suggestion of OLSEN has considerable merit and would seem to account quite well for the observed resistivities. Our primary aim has been to lend semiquantitative justification to Olsen's mechanism. To do this and maintain a clear physical picture throughout we have deliberately turned back the clock and have resorted to a most elementary approach akin to that of Nordheim. The success of that line of reasoning in heralding the results of DINGLE allows perhaps some optimism. We make, however, no claims of accuracy or rigour.

The extension of Dingle's treatment so as to include within its framework Olsen's mechanism is beset with severe mathematical difficulties. Since much available data has been obtained on polyvalent metals whose Fermi surfaces surely deviate radically from the spherical form assumed by FUCHS and DINGLE, further refinements of that theory should probably aim at removing this restriction. After that has been accomplished additional complicating mechanisms might be introduced. What we believe to have shown is that Olsen's mechanism must ultimately be included in any careful theory.

II. Estimate of Olsen's Resistivity Mechanism

The dominant resistivity mechanisms in thin wires are:

1) Bulk mechanisms; (a) impurity scattering – m.f.p. $= l_0$
(b) phonon scattering – m.f.p. $= l_i$.

2) Surface mechanisms; (a) direct surface scattering – m.f.p. $= l_s$
(b) phonon-surface scattering – m.f.p. $= l_{ps}$.

We assume *ad hoc* that the following is valid

$$\frac{1}{l_W} = \frac{1}{l_0} + \frac{1}{l_i} + \frac{1}{l_s} + \frac{1}{l_{ps}} . \tag{6}$$

It is patently obvious that Equation (6) cannot be correct. Quite apart from the usual criticisms that are leveled against Matthiessen's rule, we note that mechanism 2b is not statistically independent of mechanisms 1b and 2a. But, as we remarked before, we lay no claim to rigour here.

We concern ourselves now with the calculation of l_{ps} and restrict the discussion to the limit $l_i \gg r$. We assume that the differential scattering cross section for phonon scattering is a constant for $0 < \theta \leqslant \theta_{\max} = T/\theta_D$, and is zero for $\theta > \theta_{\max}$. If we neglect Umklapp processes then a, the average distance traveled between phonon collisions can be related to the bulk resistivity as follows

$$\left.\begin{aligned} \varphi_i(T) &= (mv_F/ne^2)\ (1/a)\ \langle(1-\cos\theta)\rangle \\ &= (\mathrm{A}/4a)\ (T/\theta_D)^2;\ T \ll \theta_D \end{aligned}\right\} \tag{7}$$

Thus we obtain

$$a = [A/4\,\varrho_i(T)]\ (T/\theta_D)^2 \tag{8}$$

Consider now an electron moving initially along the wire axis. The probability that this electron reaches the surface through multiple small angle scattering after having traversed a distance $\bar{x}$ along the wire is given by[10])

$$H(\bar{x}) = (3/4\,\pi)^{1/2}\ w(\bar{x})^{-3/2} \exp\left[-\frac{3}{4}\ w^2\ r^2\ (\bar{x})^{-3}\right] \tag{9}$$

where

$$w = [2\ a/\pi\theta^4_{\max}]^{1/2}.$$

We now define l_{ps} as that distance $\bar{x}_0$ for which the probability that the electron has reached the surface is a maximum. This definition is by no means the only acceptable one. For example, one could equally well let l_{ps} correspond to some fixed value of $H(\bar{x})$, say $1/e$. The ambiguity does not appear to us a serious objection; the choice we have made is, at least, reasonable on intuitive grounds. Another objection might be that the m.f.p. is actually the path traversed by the electron moving along its trajectory until it strikes the surface. We have taken, however, the projection of this trajectory along the wire axis. In the limit $l_i \gg r$, to which we are restricting this discussion, these two lengths are the same.

We shall now assume that the distance l_{ps} calculated in this fashion applies to all electrons, also those not moving initially along the axis of the wire. In fact, one can show that in the limit $l_i \gg r$

$$\frac{1}{l_2(\psi)} = \frac{1}{l_{2a}(\psi)} + \frac{1}{l_{2b}(0)} \tag{10}$$

where ψ is the angle between the initial direction of motion of the electron and the axis of the wire.

We maximize Equation (9) with respect to x and obtain

$$\bar{x}_0 = l_{ps} = [2\ ar^2/\pi\theta^4_{\max}]^{1/3} = [Ar^2\theta_D^2/2\,\pi\,\varrho_i(T)\,T^2]^{1/3}. \tag{11}$$

With the aid if Equation (3) and (6) we have

$$\varrho_W = \varrho_i(T) + \varrho_0 + \varrho_s + \varrho_{ps}(r, T) \tag{12}$$

where

$$\varrho_{ps}(r, T) = (2\,\pi\,A^2)^{1/3}\ (T/\theta_D)^{2/3}\ [\varrho_i(T)]^{1/3}(r)^{-2/3} \tag{13}$$

is a *size and temperature dependent contribution to the resistivity.*

III. Comparison with Experiment

Our result, given by Equation (13), allows direct comparison with the results of OLSEN and ANDREW. We call attention here to the fact that *Equation (13) contains no adjustable parameters whatever.* The quantity A for indium[8]) and mercury[9]) is approximately equal to 2×10^{-11} ohm-cm^2 for both metals. The ideal bulk resistivities are obtained most conveniently by extrapolating the resistivities of the thickest wires, whose surface effects are negligible, to $T = 0°\,\mathrm{K}$, thereby obtaining the bulk residual resistivities. By a simple process of subtraction one now finds $\varrho_i(T)$, the lowest curves of Figures 1 and 2. The wire radius, r, is also known.

The DEBYE characteristic temperature of indium has been determined quite accurately[11]). Below $3°\,\mathrm{K}$ θ_D is constant at $109°\,\mathrm{K}$; between $3°\,\mathrm{K}$ and about $5°\,\mathrm{K}$ it falls, and at $4{,}2°\,\mathrm{K}$ its value is $106°\,\mathrm{K}$. The Debye temperature of mercury is unfortunately not so well known. BLACKMAN[12]) gives $\theta_D = 90°\,\mathrm{K}$, GERRITSEN[13]) quotes $\theta_D = 69°\,\mathrm{K}$, KLEMENS[14]) seems to favor $\theta_D = 60°\,\mathrm{K}$, and HULM[15]), who is particularly concerned with the low temperature region, offers $\theta_D = 37°\,\mathrm{K}$. We arbitrarily select yet another value which is intermediate of the extremes but leans more toward Hulm's result, namely $\theta_D(\mathrm{Hg}) = 50°\,\mathrm{K}$. Should subsequent determinations prove this choice completely unrealistic the error is easily corrected.

If we now take the difference between the ordinate of a particular curve of Figure 1 and the ordinate of the curve marked In-6,2 (the ideal resistivity of the bulk) we have the experimental value of ϱ_{ps}. Similarly for mercury, using here Figure 2 and the curve marked Hg-1 as the bulk ideal resistivity curve. The experimental values of ϱ_{ps} are shown by solid curves in Figure 3 and 4, and the calculated curves are shown dashed. In Figures 4 the arrows indicate that temperature at which $l_i = r$; since our derivation applies only in the limit $l_i \gg r$, comparison of Equation (13) with the results of ANDREW should be limited to the temperature region below $2{,}5°\,\mathrm{K}$.

The excellent qualitative agreement between the calculated and experimental results is immediately apparent. Moreover, in view of the absence of any adjustable parameters whatever, the order of magnitude agreement is also gratifying. It is significant that the calculated results consistently exceed the experimental by factors of about 3 in the case of indium and of about 2 in the case of mercury. We shall presently return to discuss and interpret this discrepancy which, we think, has an interesting significance and sheds light on the conduction properties of the bulk metal.

Figures 5 and 6 provide a comparison of the observed and predicted *size dependence* of ϱ_{ps}. According to Equation (13) ϱ_{ps} should be propor-

tional to $r^{-2/3}$ at a fixed temperature. Consequently, if, for some given temperatures, we plot ϱ_{ps} against $r^{-2/3}$ we should obtain a family of straight lines emanating from the origin. In the case of indium, with a fair number of experimental points available, we see that these do indeed form a family of straight lines as predicted. In the case of the mercury wires we again find good agreement with the $r^{-2/3}$ relation at 2,0° K. At 2,5 K the point corresponding to the wire of largest diameter falls below the straight line. This is the deviation from our theoretical result which is to be expected. As the wire diameter becomes comparable to the bulk m.f.p. the theoretical result, predicated on the assumption that $l_i \gg r$, must overestimate surface effects. The deviation of the experimental points from the straight line at 3·0° K comes as no surprise. We conclude that in the region of validity of the theory, the data that are available are in excellent agreement with the predicted size variation of ϱ_{ps}.

Before comparing the predicted and observed temperature dependence we turn our attention to the systematic overestimate of the theory for

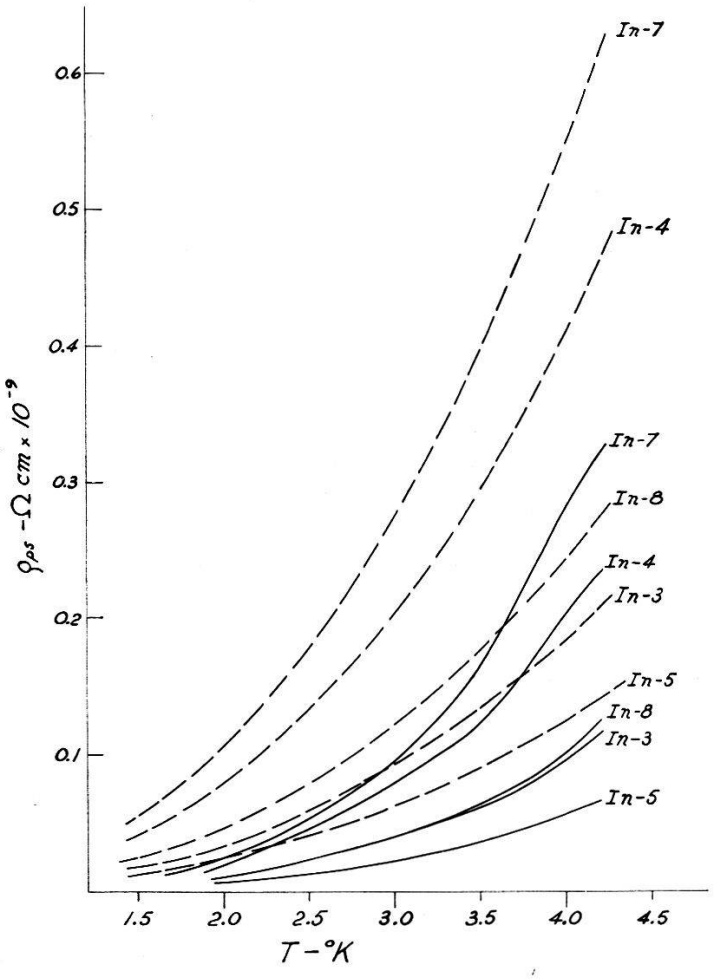

Fig. 3

The observed (solid lines) and calculated (dashed lines) resistivities of thin indium wires due to phonon-surface scattering.

ϱ_{ps} to which have already called attention. In retrospect we recognize that this is indeed what we might have anticipated once we realize that *not all phonon scattering leads to small angle events*. It is well known that Umklapp and Normal processes contribute to the ideal resistivity of a metal, with the former making perhaps the dominant contribution. Umklapp processes are generally large angle events, and the presence of the surfaces does not enhance the effectiveness of Umklapp scattering. Our theoretical results are too large because we have used the total ideal resistivity $\varrho_i(T)$ in Equation (13), whereas we should only use that part attributable to Normal scattering. Thus, we should write $\varrho_i^N(T)$ in Equation (13) rather than the total ideal resistivity.

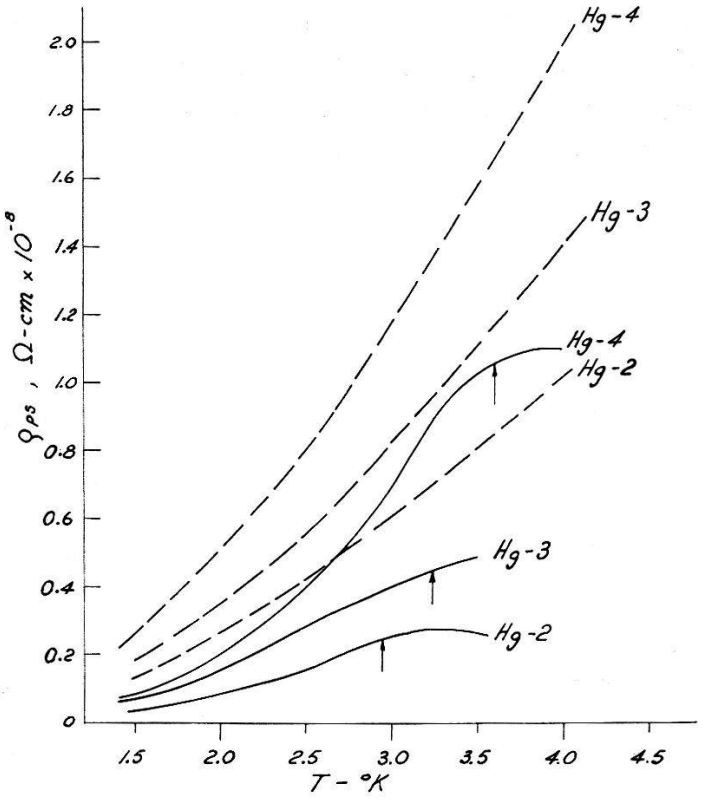

Fig. 4

The observed (solid lines) and calculated (dashed lines) resistivities of thin mercury wires due to phonon-surface scattering.

To bring calculated and observed values of ϱ_{ps} into numerical agreement we must set

$$\text{for indium:} \quad \varrho_i^N(T) \simeq \varrho_i(T)/27; \quad \frac{\varrho_i^N(T)}{\varrho_i^U(T)} \simeq 0{,}04$$

$$\text{for mercury:} \quad \varrho_i^N(T) \simeq \varrho_i(T)/8; \quad \frac{\varrho_i^N(T)}{\varrho_i^U(T)} \simeq 0{,}14.$$

These ratios for the resistivities due to Normal and Umklapp events are most reasonable[16]). Indeed, we believe that one of the most interesting and important results of an investigation such as Olsen's may be that it allows an estimate of the Umklapp resistivity.

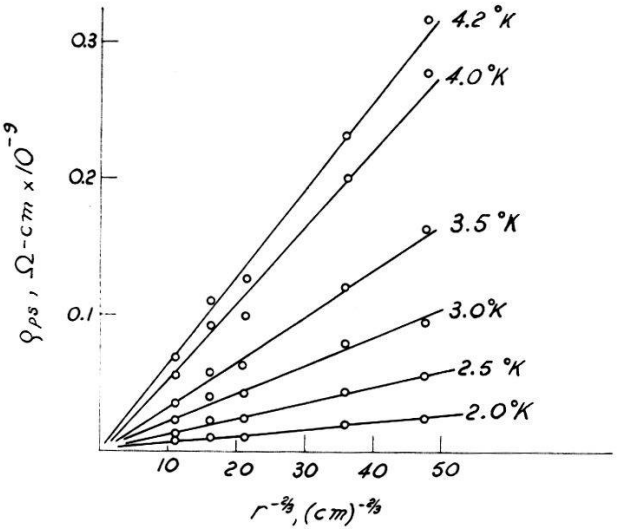

Fig. 5

The observed (0) and predicted (straight lines) size dependence of the resistivity of thin indium wires due to phonon-surface scattering.

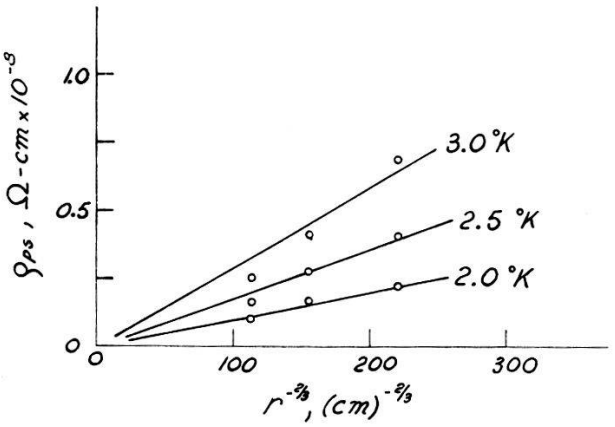

Fig. 6

The observed (0) and predicted (straight lines) size dependence of the resistivity of thin mercury wires due to phonon-surface scattering.

If the argument of the preceding paragraph is accepted, a comparison of the predicted and observed temperature dependence is now slightly complicated. We cannot make use of the observed temperature dependence of the ideal resistivity of the bulk samples; instead, we need to know the temperature dependence of $\varrho_i^N(T)$, that portion of the ideal resistivity

arising from Normal scattering only. According to Bloch theory[17] $\varrho_i^N(T)$ is proportional to T^5. That theory is, however, based on the free electron model – on all the assumptions and approximations implicit therein – certainly an extreme and untenable oversimplification of the true state of affairs. At the same time, we recognize that we have employed this very same model here ourselves, and the use of the results of the Bloch theory is, at least, consistent. Moreover, a T^5 law appears to be approximately valid in many cases and is, probably, not far off the mark.

If we then accept the T^5 law it follows from Equation (13) – now modified by the replacement of $\varrho_i(T)$ by $\varrho_i^N(T)$ – that ϱ_{ps} should be proportional to $T^{2.33}$. The slopes of the straight lines of Figures 5 and 6 are proportional to the temperature dependent factors appearing in Equation (13). Consequently, a plot of these slopes as functions of the temperature should reval the average experimental temperature dependence. Such logarithmic plots are shown in Figures 7 and 8 together with straight lines

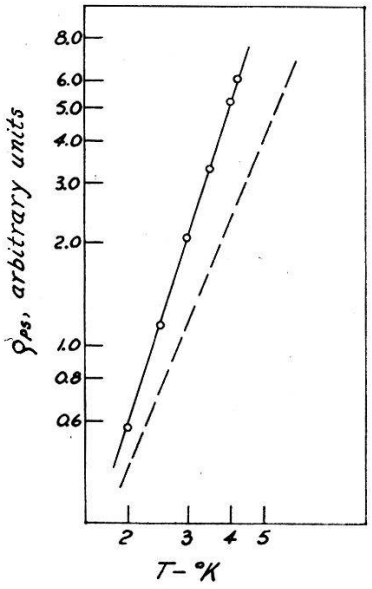

Fig. 7

The average temperature dependence of the resistivity of thin indium wires due to phonon-surface scattering. The predicted slope is shown by the dashed line.

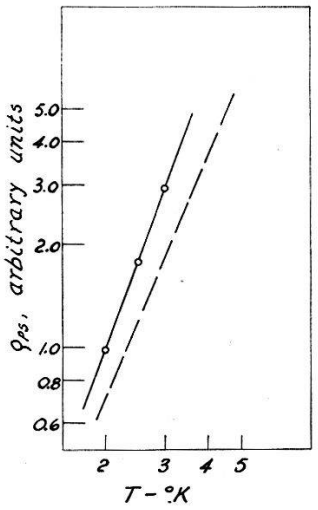

Fig. 8

The average temperature dependence of the resistivity of thin mercury wires due to phonon-surface scattering. The predicted slope is shown by the dashed line.

(dashed) corresponding to the theoretical slope of 2·33. For the indium samples $\log \varrho_{ps}$ *versus* $\log T$ yields a straight line with a slope of nearly three; for the mercury wires a slope of about 2·6 is indicated. (If the points of Figures 6 are plotted individually one obtains straight lines whose slopes are 2·3, 2·5, and 2·9.) The expression for ϱ_{ps} which we derived

does not quite agree with the observed temperature dependence. However, the discrepancy is sufficiently small to suggest that we are on the right track. It is very likely that one or another assumption which we were forced to make causes the result to display a somewhat weaker temperature dependence than it should.

IV. Conclusion

The results presented in the previous sections suggest that the explanation for the failure of Matthiessen's rule in thin wires originally proposed by OLSEN is indeed quite plausible.

We have obtained an analytic expression for the phonon-surface resistivity which may be brought into excellent numerical agreement with existing data on indium and mercury wires provided a reasonable ratio of Normal-to-Umklapp scattering is postulated. The predicted size dependence of the phonon-surface resistivity is in excellent agreement with observation. The predicted temperature dependence is as $T^{2.33}$, and the experimental results show dependences ranging from $T^{2.3}$ to $T^{3.0}$, the more rapid temperature variation being favored.

The assumptions which we have made have been numerous and rather difficult – not to say impossible – to justify. We believe that the best justification for our procedure rests in its ability to yield a simple analytic expression which allows comparison with experiment. The method obviates long and tedious numerical computations which often obscure insight into the physical significance of the important mechanisms and whose results are generally not so readily compared to experimental data. As this manuscript was being prepared we received a preprint of a paper (published as companion to the present paper) by LÜTHI and WYDER[18], who attacked the same problem from a rather different avenue. They calculated the m.f.p. of free electrons in a thin wire by means of a Monte-Carlo calculation. This approach has the advantage that it avoids one of the assumptions which we were forced to make, namely that involving the statistical independence of the several relaxation mechanisms. Moreover, their numerical approach is not limited to $l_i \gg r$, as is our calculation.

LÜTHI and WYDER seem to reproduce the experimental results reasonably well. Unfortunately, the numerical approach does not allow for the same detailed comparison with the data of OLSEN and ANDREW as we could give (see Figures 5, 6, 7, and 8). The qualitative agreement of their results and ours suggests that the strengthening of one of the weakest links of our argument does not appear to change the final result very much. It is surprising, however, that LÜTHI and WYDER obtain good

numerical agreement under the assumption that Umklapp processes may be neglected altogether. This is in sharp contrast to our view that Umklapp processes are approximately as important in indium and mercury as they are in most other metals.

Quite apart from the approximations and assumptions we have indulged and which have been stated explicitly, a number of others are implicit in this as well as in the work of FUCHS, DINGLE, and of LÜTHI and WYDER.

First, we have not considered the possible effects of localized surface (TAMM) states. Very little is known of surface states in metals although in recent years investigations in this field have been initiated[19].

Secondly, we have assumed throughout, as did all other workers, that a continuum of states in momentum space is available to electrons and phonons. It is clear, however, that in a thin wire the number of vibrational modes with wave vector normal to the wire axis is $3N_A$, where N_A is the number of atoms in a cross sectional area of the wire. For very thin wires this number may be sufficiently small so that at low temperatures the energy difference between neighboring vibrational modes with wave vector normal to the wire axis is of order kT. In that event the usual description (density of states, etc.) for electron-phonon scattering must fail[20]). Size effects attributable to the discreteness of the phonon spectrum have been observed in other experiments[21]). Of course, the same considerations also apply to the spectrum of electron energies. Consequently, a treatment which assumes continuous electron and phonon spectra *a priori* cannot be correct in the limit of thin wires and low temperatures.

In conclusion we believe that the success which we have attained, as well as that reported by LÜTHI and WYDER, establishes Olsen's mechanism as one of the dominant ones in thin samples. In contrast to LÜTHI and WYDER, who neglect Umklapp entirely, we believe that careful experiments on thin wires provide a means of separating the Normal and Umklapp contributions to the total resistivity in a metal.

V. Acknowledgements

A portion of the material presented here formed part of the dissertation submitted by one of us (H. G. S.) to Michigan State University in partial fulfillment of the requirements for the M. S. degree in Physics. The work was continued and the paper prepared while the other (F. J. B.) held an N. S. F. post-doctoral fellowship at the Clarendon Laboratory, Oxford. The hospitality of Prof. W. E. LAMB, Jr. and of PROF. B. BLEANEY is gratefully acknowledged. A preliminary report of the work was presented in August, 1959, at the Solid State Conference in Melbourne, Australia.

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