CONDUCTIVITY OF THIN METALLIC WIRES

Thesis for the Degree of M. S.

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Helmut Gerhard Satz

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By

Helmut Gerhard Satz

AN ABSTRACT

Submitted to the College of Science and Arts
Michigan State University of Agriculture and
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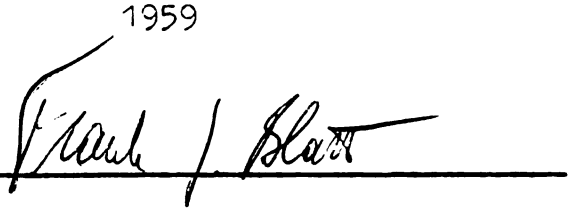
Approved 

ABSTRACT

Measurements by Olsen[+)] on thin indium wires at low temperatures have demonstrated that not only the residual but also the temperature dependent part of the resistivity increases as the wire diameter decreases. It was suggested by Olsen that small angle phonon scattering, which may take electrons to the surface, where they suffer diffuse scattering, might give rise to the observed effect. Since an exact solution of the transport equation in this case is beset with nearly insurmountable difficulties, we have resorted to an extremely crude analysis similar to that employed by Nordheim[++)]. Two mechanisms are considered: 1) Electron-phonon scattering, which takes electrons to the surface in a time shorter than τ_{ph}^{b}, where τ_{ph}^{b} is the electron-phonon relaxation time in the bulk. 2) Electron-electron scattering, which, although of no consequence in the bulk, may also contribute to the resistivity of a thin wire by bringing carriers to the surface. Both processes lead to a size and temperature dependent contribution to the total resistivity of thin wires, although in the temperature region considered here the size dependence of the electron-electron effect is very weak, and thus this process could account for at most a small part of the observed resistivity.

+): Olsen, J. L., Helv. Phys. Acta, 31, 713 (1958)

++): Nordheim, L., Act. Sci. et Ind., No.131 (Paris:Hermann)

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I. INTRODUCTION

If one attempts to explain the electrical properties of metals on the basis of an essentially free electron gas obeying Fermi-Dirac statistics (Sommerfeld theory), then the electrical conductivity is obtained in terms of the electronic "mean free path" ℓ as

$$\sigma = \left[\frac{ne^2}{m\bar{v}}\right]\ell$$

where e and n are the electronic charge and the number of electrons per unit volume, respectively; $\bar{v}$ is the velocity of an electron at the surface of the Fermi distribution, and m the mass (or effective mass) of the electron. Since $\bar{v}$ remains quite constant over a very wide range of temperature from $0^{\circ}K$ upward (the electrons forming a strongly degenerate Fermi gas), the above relation gives the conductivity, aside from constants, in terms of only the mean free path.[1)]

At normal temperatures this mean free path is very much shorter than any dimension of a specimen, and hence the conductivity in that temperature region is independent of the size and shape of the conductor. As the temperature is decreased, however, the mean free path in a pure metal increases, due to a diminished number of thermal vibrations (phonons) present at lower temperatures, and hence for thin wires and films at very low temperatures the dimensions of the conductor may be comparable to or smaller than the mean free path, thus suggesting that under these circumstances

there should be a dependence of the conductivity on the geometrical shape of the conductor. Experimentally it is indeed found[2] that at very low temperatures the conductivity of thin wires and films decreases with decreasing diameter or thickness. This result can be interpreted as an additional resistivity mechanism arising from an increased importance of electron collisions with the surface of the conductor, assuming these collisions to be at least partially diffuse. In calculating the low temperature conductivity of thin wires or films the problem then arises as to how these additional surface effects are to be treated.

The major theoretical investigations of this question were carried out by Fuchs[3] and Dingle[4]; both authors give critical evaluations of the work done previous to theirs. A brief review of Fuchs' method for thin films will now be given here; Dingle's work consists essentially in extending this procedure to a cylindrical geometry.

In Fuchs' treatment the following main assumptions are made:

1) Electron scattering at the surface is completely diffuse; i.e., an electron is scattered from the surface into any solid angle with equal probability, independent of its initial direction of motion. (Fuchs also treats the case of partially diffuse, partially specular scattering; experimental results[5] seem to indicate, however, that such scattering is largely diffuse, and hence only this case will be considered here).

2) In the bulk material the probability per unit time that an electron is scattered through an angle Θ is independent of Θ ; this probability may, however, be a function of the electron energy.

These assumptions state in effect that any collision suffered by an electron will completely randomize its momentum, independent of the initial direction of motion.

A transport equation for the disturbed distribution function $f(\vec{v}, z)$ can then be written, in which the integral operator for scattering in the bulk is replaced by a "relaxation" term involving a relaxation time τ ; this is justified by assumption 2) above. (For the geometry, see Figure I-1). The non-equilibrium distribution function $f(\vec{v}, z)$ is now expressed in terms of the equilibrium (zero electric field) distribution function $f_0(|\vec{v}|)$ and a small perturbation term $f_1(\vec{v}, z)$:

$$f(\vec{v}, z) = f_0(v) + f_1(\vec{v}, z)$$

With this, the transport equation becomes, to first order, a partial differential equation for $f_1(\vec{v}, z)$:

$$\frac{\partial f_1}{\partial z} + \frac{f_1}{\tau v_z} = \frac{eE}{m v_z} \frac{\partial f_0}{\partial v_x} \qquad \text{(I-1)}$$

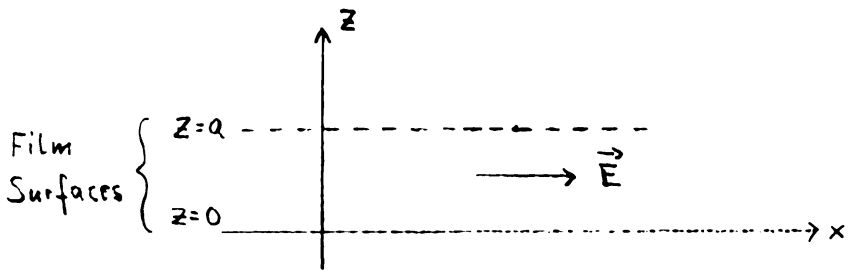

Figure I-1)

Equation (I-1) is then solved, subject to the following boundary conditions:

At $z = 0$, $f_1(\vec{v}, z)$ is independent of the direction of $\vec{v}$ for all $v_z > 0$, and at $z = a$, $f_1(\vec{v}, z)$ is independent of the direction of $\vec{v}$ for all $v_z < 0$; these conditions are equivalent to the assumption of perfectly diffuse scattering at the surfaces. The solution of (I-1) is used to find the mean current density and hence the conductivity:

$$j = \sigma E = \int_0^a \left[e \iiint v_x f(\vec{v}, z)\, dv_x\, dv_y\, dv_z \right] dz$$

The results thus obtained can be written in terms of elementary functions only for the limiting cases of thick and very thin films, i.e., for $\ell \ll a$ and $\ell \gg a$, where ℓ is the mean free path and a the film thickness. The results are given below, together with the corresponding expressions for wires, as found by Dingle[4]:

FILMS:

$$\frac{\sigma_b}{\sigma} = 1 + \frac{3}{8K} \quad \cdots\cdots\cdots\cdots\cdots \quad K \gg 1 \qquad \text{(I-2)}$$

$$\frac{\sigma_b}{\sigma} = \frac{4}{3K}\left[1/\log\left(\frac{1}{K}\right)\right] \quad \cdots\cdots\cdots \quad K \ll 1 \qquad \text{(I-3)}$$

where $K = a/\ell$; σ_b and a are bulk conductivity and film thickness, respectively.

WIRES:

$$\frac{\sigma_b}{\sigma} = 1 + \frac{3}{4K} \quad \cdots\cdots\cdots\cdots\cdots \quad K \gg 1 \qquad \text{(I-4)}$$

WIRES, continued:

$$\frac{\sigma}{\sigma_0} = {}^{1}/_{K} \quad \cdots\cdots\cdots\cdots\cdots\cdots \quad K \ll 1 \qquad \text{(I-5)}$$

where $K = {}^{d}/_{\ell}$; d is the wire diameter.

These results can now be compared with experiment in two different fashions:

1) The conductivity σ may be measured experimentally at one or more fixed temperatures for specimens of varying thicknesses; in other words, K is varied by choosing various values of a or d , for a fixed ℓ .

2) σ may be measured for a fixed thickness or diameter at various values of the temperature; i.e., K is varied by letting ℓ vary at a fixed a or d .

Experiments using method 1), carried out for various metals[6], gave fairly good agreement with the Fuchs-Dingle theoretical results. Experiments employing method 2) over a fairly wide temperature range have been performed only recently by Olsen[7] and do not seem to agree with the above theoretical calculations (see Figure I-2 and end of Section I.). To find a possible reason for this disagreement it will be necessary to consider in somewhat greater detail the various mechanisms responsible for the resistivity of thin wires or films.

In the bulk material there exist two types of resistance mechanisms:

a) Scattering by impurities, vacancies, dislocations, etc.; this mechanism is temperature independent and hence will give

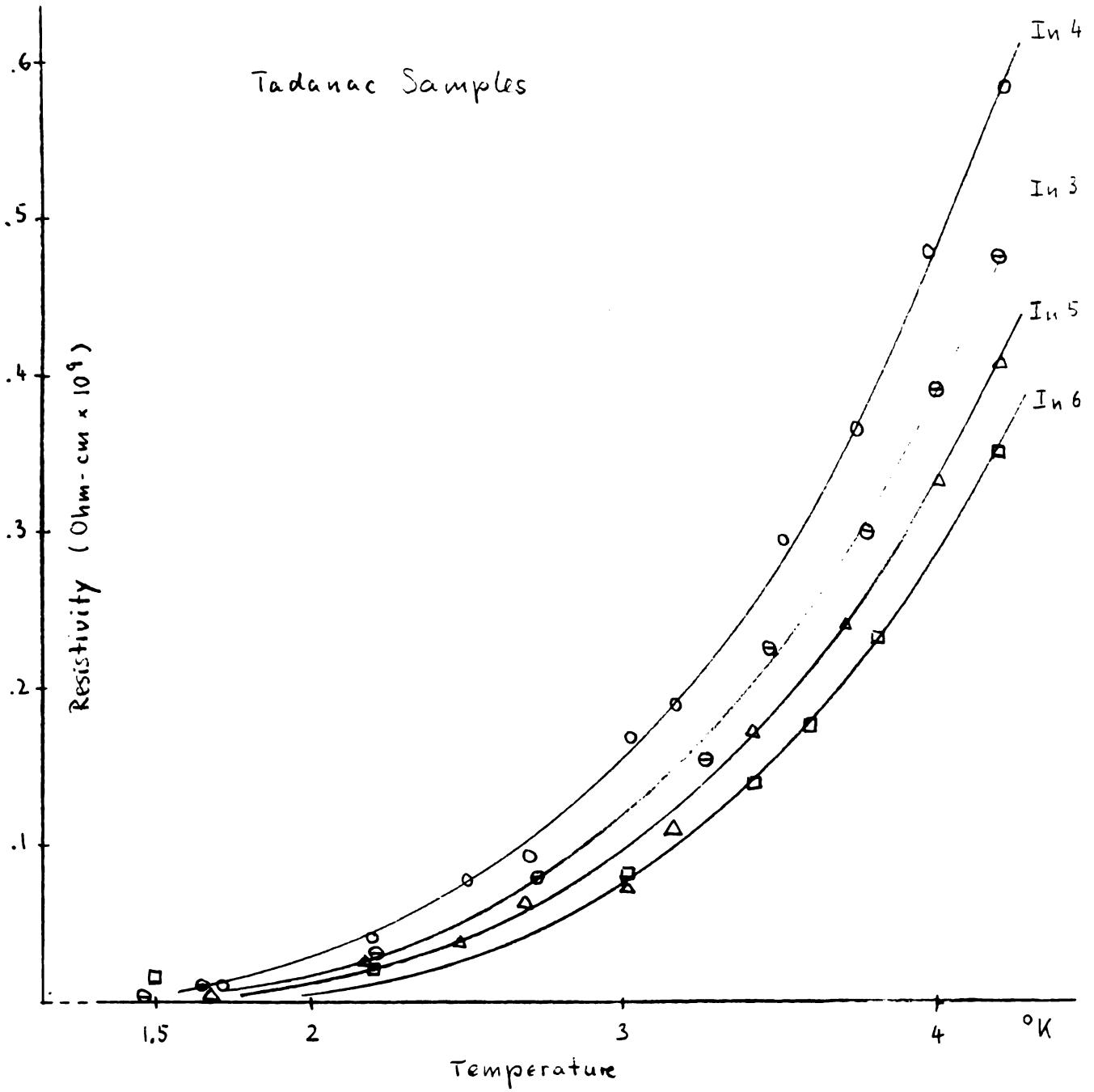

Figure I-2a

The temperature dependent resistivity of thin Indium wires: Olsen's experimental results. Tadanac Brand.

Specimens: In 3 (⊖), cross-section .311 mm
In 4 (○), cross-section .0855 mm
In 5 (△), cross-section .57 mm
In 6 (□), cross-section 2.54 mm

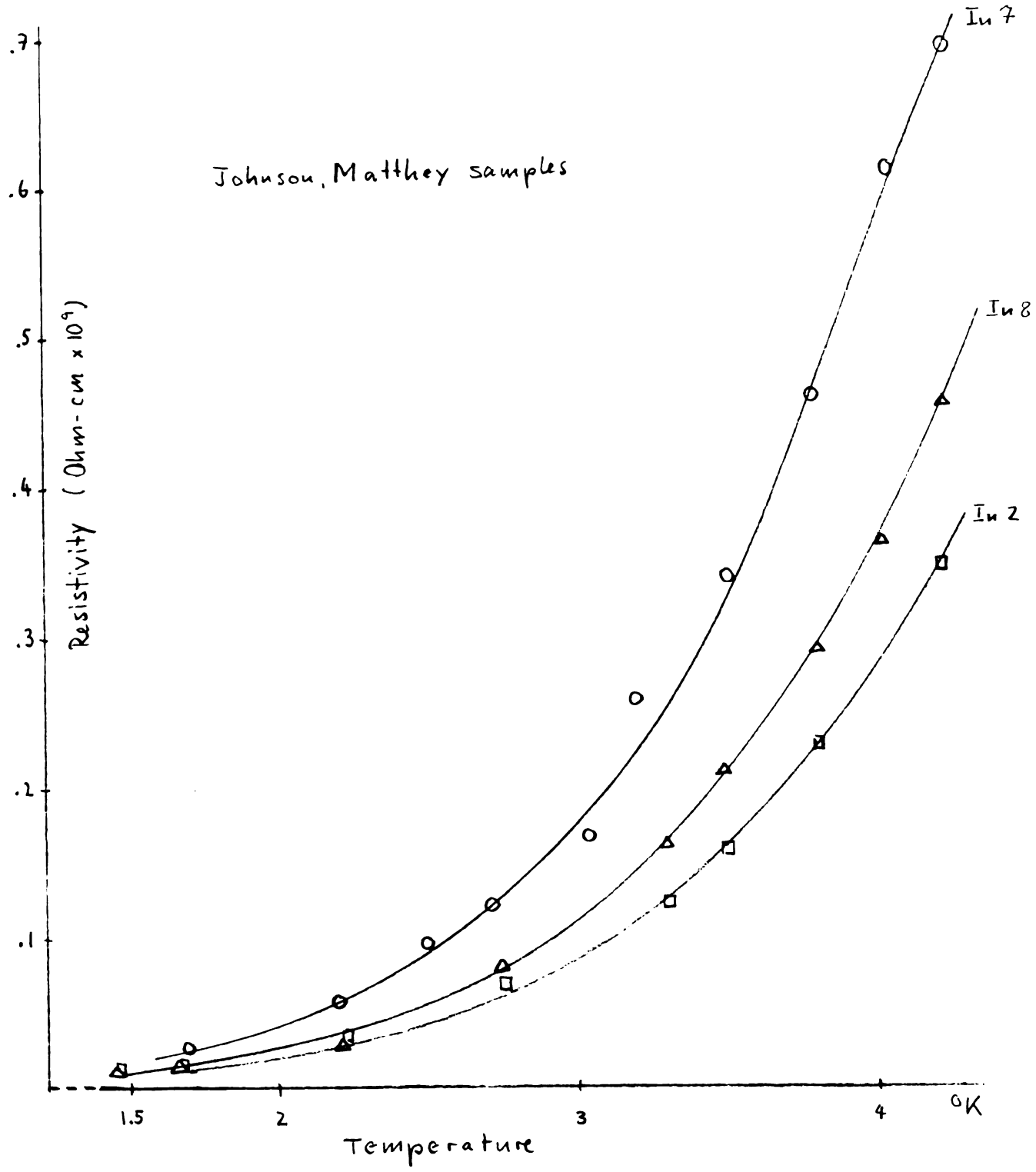

Figure I-2b

The temperature dependent resistivity of thin Indium wires: Olsen's experimental results. Johnson, Matthey samples.

Specimens: In 2 (□), cross-section 2.0 mm
In 7 (○), cross-section .06 mm
In 8 (△), cross-section .20 mm

a temperature independent contribution to the resistivity ("residual resistivity").

b) Scattering by thermal lattice vibrations (phonons); this mechanism is temperature dependent: the average angle of scattering decreases with temperature and vanishes as T approaches $0^{\circ}K$. Lattice scattering therefore gives a temperature dependent contribution to the resistivity ("ideal resistivity").

In addition to these there is electron-electron scattering. In the bulk material this effect leads (in the region of validity of Ohm's law) to no resistivity, since the total electronic momentum is conserved in such processes. In thin films or wires, however, electron-electron scattering may bring electrons, which would otherwise be scattered in the bulk, to the surface, where the electron momentum is randomized, hence giving rise to a resistive effect. Since Fuchs uses the surface only as a boundary condition on bulk resistivity processes, i.e., since $\bar{\tau}$ is a relaxation time corresponding to bulk mechanisms only, any pure surface effects, such as electron-electron-to-surface scattering, are automatically excluded.

Fuchs' method now requires the existence of a relaxation time $\bar{\tau}$ to account for all mechanisms in the bulk material. Since the two types of bulk resistance mechanisms are statistically independent and since $1/\bar{\tau}$ is proportional to the transition probability, one would obtain

$$\frac{1}{\bar{\tau}} = \frac{1}{\bar{\tau}_{resid.}} + \frac{1}{\bar{\tau}_{ideal}}$$

provided that an unambiguous relaxation time can be defined for each of the two processes. Assuming, as required by Fuchs' approach, that this can be done, one finds thus that in the bulk material a separation of the total resistivity into a sum of a temperature independent and a temperature dependent part is possible:

$$\rho_b = \rho_b^{resid} + \rho_b^{ideal} \qquad \text{(Matthiessen's Rule)}$$

Considering now the results of the theory for wires (since Olsen investigated indium wires of various diameters over a low temperature region), one finds for the resistivity (cf. equations I-4 and I-5):

For $K \gg 1$,

$$\rho_w = \rho_b + 3/4\,[\rho_b \ell / d\,]$$

$$= \rho_b^{resid.} + \rho_b^{ideal} + 3/4\,[\rho_b \ell / d\,]$$

$$= \rho_b^{resid} + \rho_b^{ideal} + \text{constant} \times (1/d)$$

and for $K \ll 1$,

$$\rho_w = \rho_b \ell / d = \text{constant} \times (1/d)$$

where $\rho_b \ell = \ell / \sigma_b = m\bar{v}/ne^2 = \text{constant.}$

The theoretical results thus predict that for thin wires Matthiessen's rule is still valid. For $K \gg 1$, the temperature variation of ρ_w is the same as that of the bulk material; for $K \ll 1$, ρ_w becomes a constant, intuitively on the grounds that all free paths now start and end on the surface of the wire, whence the resistivity should be due largely

to temperature independent surface effects. At any rate, in both limits the temperature dependence of the wire resistivity is independent of surface effects, i.e., independent of the wire diameter.

Olsen's results[7], however, indicate a pronounced dependence of the temperature variation of ρ_w on the diameter of the wire (cf. Figure I-2). He finds that the resistivity ρ_w increases faster with temperature the thinner the wire. Thus, Matthiessen's rule here is not strictly valid: there is no ideal resistivity, since effects due to the geometrical configuration of the conductor cannot be separated from thermal effects.

Olsen suggests the following explanation: At low temperatures an electron-phonon scattering event produces only a very small deviation of the electron path, and hence in the bulk material many collisions are necessary before this mechanism randomizes the electron momentum; in the case of thin wires, however, a much smaller number of collisions may bring the electron to the surface, where its momentum is randomized - thus the effective temperature dependent mean free path is shorter in the wire, i.e., the resistivity is greater, the thinner the wire.

We shall attempt in Section III. to use a similar argument in order to obtain semi-quantitative results to account for the observed deviation from Matthiessen's rule.

II. OUTLINE OF THE STRICT TRANSPORT THEORY ANALYSIS.[+)]

Since the Fuchs treatment, in spite of fairly good agreement with experiments performed at constant temperature for wires of various diameters (method 1., p. 5), failed to predict the correct experimental behaviour when the temperature variations for wires of different radii were compared (method 2., p. 4), we shall attempt to employ a more general transport theory treatment.

It is recalled that Fuchs' method consisted essentially of setting up the transport equation, eliminating the integral scattering operator by assuming the existence of a relaxation time (for all bulk mechanisms), then substituting a perturbation expansion $f = f_0 + f_1$ for the disturbed distribution function f, and finally solving the partial differential equation for f_1, subject to the boundary

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+) : Electron-electron-to-surface effects will be excluded here from the outset, for two reasons: (1) Our objective in this section is essentially to analyse the possibility of a transport theory treatment even if only impurity etc., phonon and simple surface scattering mechanisms are present; (2) Should a strict transport theory approach prove to be feasible, electron-electron effects could be included in a somewhat more general treatment.[8)] - In the phenomenological approach to be developed in Section III., we shall consider these electron-electron-to-surface effects along with the other possible resistance mechanisms.

conditions of totally diffuse scattering at the film surfaces. Mathematically then, assumption 1) above (p.2) provided the boundary conditions for the transport equation, assumption 2) eliminated the integral operator, thereby reducing an integro-differential equation to a partial differential equation. It was further assumed implicitly that a perturbation expansion of f is valid; this is physically reasonable and will continue to be maintained. Assumption 1), regarding the diffuseness of the surface scattering, is not essential; one could assume partially specular reflection by applying somewhat altered boundary conditions.[3] However, completely diffuse scattering seems to be fairly well justified experimentally[5], and therefore this assumption will also be maintained. Assumption 2) will, for the time, be dropped, since there seems to be no a priori justification for it (see below).

The general transport equation then becomes

$$-v_z \frac{\partial f}{\partial z} + \frac{eE}{m} \frac{\partial f}{\partial v_x} = \hat{S} f \qquad \text{(II-1)}$$

where again the film surfaces are $z = 0$ and $z = a$; the electric field is along the x-axis. $f = f(\vec{v}, z)$, and $\hat{S}$ is the integral scattering operator, such that $\hat{S}f = \left.\frac{\partial f}{\partial t}\right]_{scatter.}$, i.e.,

$$\hat{S}f = \int f(\vec{v}', z) P(\vec{v}', \vec{v})\, d\vec{v}' - f(\vec{v}, z) \int P(\vec{v}, \vec{v}')\, d\vec{v}' \qquad \text{(II-2)}$$

Here $P(\vec{v}', v) = P(\vec{v}, \vec{v}')$ is the transition probability per unit time from state $\vec{v}'$ to $\vec{v}$ and vice versa. Introducing now the perturbation expansion for f, equation (II-2) becomes

$$\hat{S}f = \hat{S}f_1 = -f_1(\vec{v},z)\int\left\{1-\frac{f_1(\vec{v}',z)}{f_1(\vec{v},z)}\right\}P(\vec{v},\vec{v}')\,d\vec{v}' \qquad \text{(II-3)}$$

If one now again assumes that in the bulk material there exist two statistically independent scattering mechanisms, one due to stationary scattering centers, the other due to phonons, then

$$P = P_I + P_T$$

where P_I is the impurity and P_T the phonon scattering transition probability. With this substitution equation (II-3) separates into

$$\hat{S}f = \hat{S}f_1 = -f_1\left[\int\left\{1-f_1'/f_1\right\}P_I\,d\vec{v}' + \int\left\{1-f_1'/f_1\right\}P_T\,d\vec{v}'\right] \qquad \text{(II-4)}$$

where $f_1' = f_1(\vec{v}',z)$, etc.

It is noted that this possible separation, i.e., the statistical independence of the two types of resistance mechanisms, does <u>not</u> imply matthiessen's rule; the requirements for the validity of the latter go considerably further; namely, it has to be required[9] above and beyond (II-4) that

1) unambiguous relaxation times can be defined for both types of processes;
2) the ratio of these relaxation times is independent of $\vec{v}$.

Requirements 1) and 2) are contained in the assumption made in the Fuchs-Dingle treatment, that a relaxation time τ can be defined for the total of all resistivity mechanisms, independent of the scattering angle and of the electron's initial direction of motion.

In the general equation (II-4) neither of these two conditions is necessarily fulfilled. It will, therefore, be necessary to investigate more explicit expressions for P_I and P_T .

Assuming static imperfections, P_I can be written as[10]

$$P_I(\vec{v},\vec{v}') = P_I(\theta) = \left[\vec{v}/\Omega\right] I(\theta) \qquad \text{(II-5)}$$

Here θ is the angle between $\vec{v}$ and $\vec{v}'$, the initial and final velocities of the electron; Ω is the atomic volume (assuming one conduction electron per atom), and $I(\theta)$ is the scattering cross-section of the particular type of imperfection.

Similarly P_T can be written[11]

$$P_T(\vec{v},\vec{v}') = P_T(\theta) = a_1 C\left[\frac{q^2 N_q}{\omega(\vec{q})}\right]\delta(\epsilon_{\vec{k}+\vec{q}} - \epsilon_{\vec{k}} - \hbar\omega(\vec{q})) \qquad \text{(II-6)}$$

$$+ a_2 C\left[\frac{q^2(N_q+1)}{\omega(\vec{q})}\right]\delta(\epsilon_{\vec{k}-\vec{q}} - \epsilon_{\vec{k}} + \hbar\omega(\vec{q}))$$

Here a_1 and a_2 are constants; C is the electron-phonon coupling "constant"; $\vec{q}$ is the phonon wave vector with the corresponding frequency $\omega(\vec{q})$; N_q is the number of phonons with wave vectors $\vec{q}$; finally, $\epsilon_{\vec{k}}$ is the electron energy, corresponding to an electron with a wave vector $\vec{k}$ (proportional to $\vec{v}$). The first term of (II-6) corresponds to absorption, the second to emission of a phonon.

From these expressions it is clear that in general both P_I and P_T are dependent on $\vec{v}$, and moreover, that this dependence in general is different for P_I and P_T : in P_I

it is temperature independent but varies with the type of impurity, while in P_T it is strongly temperature dependent, in particular through the dependence of N_q on temperature (Planck's distribution). Moreover, a relaxation time for electron-phonon scattering can generally not be defined for temperatures much below the Debye temperature[12].

In a correct analysis it is therefore not permissible to define a relaxation time including all bulk processes at the outset, since requirements 1) and 2) (p.11) are probably not satisfied.

In order to solve the problem exactly one would thus have to solve, without any further major approximations, the transport equation as it stands:

$$-v_z \frac{\partial}{\partial z} f_1(\vec{v},z) + \frac{eE}{m} \frac{\partial}{\partial v_x} f_0(v) = -f_1(\vec{v},z) \times \tag{II-7}$$

$$\times \left\{ \int \left[1 - \frac{f_1(\vec{v}',z)}{f_1(\vec{v},z)} \right] P_I(\vec{v},\vec{v}')\, d\vec{v}' + \int \left[1 - \frac{f_1(\vec{v}',z)}{f_1(\vec{v},z)} \right] P_T(\vec{v},\vec{v}')\, d\vec{v}' \right\}$$

where P_I and P_T are given by (II-5) and (II-6); equation (II-7) is then subject to the appropriate boundary conditions of completely diffuse scattering at the surfaces.

The strict analysis will not be carried beyond this point, for the following reasons:

Equation (II-7) is an integro-differential equation in which the kernel of the integral operator, among other things, is dependent on the boundary conditions. Its solution, if at all possible, would be mathematically very difficult, if no further simplifying assumptions can be made. A solution

has been obtained by Sondheimer[13], but under the very special assumption that $P_T(\theta) = a \cos\theta$.

What will therefore be attempted instead is the following. Abandoning a rigorous mathematical procedure based on transport theory, we shall try to construct a very crude picture of the processes involved, based on simple physical models, and try to see whether it is possible to obtain results giving at least rough overall agreement with experimental results. That such an approach may not be totally worthless is perhaps supported by the fact that a very crude "kinetic" treatment of Nordheim[14] gives results which for thin wires agree to within 5%[15] over the entire range of K (i.e., of various diameters at a fixed temperature) with the "exact" results of Dingle's treatment. One may therefore hope that a crude treatment based on simple physical models will at least give some insight into the role played by several scattering mechansims not considered by Fuchs or Dingle.

III. KINETIC APPROXIMATION.

At the outset we shall classify the various resistivity mechanisms present in thin wires and films as follows:

A) Bulk mechanisms (momentum randomized without surface interaction)

1) Scattering by lattice imperfections --- temperature independent.

2) Scattering by phonons --- temperature dependent.

B) Surface mechanisms (momentum randomized by completely diffuse scattering at the surfaces)

3) Simple surface scattering --- temperature independent.

4) Small angle phonon scatterings which bring the electron to the surface --- temperature dependent.

5) Electron-electron collisions which bring the electron to the surface --- temperature dependent.

The bulk processes are assumed to be known and are given in terms of mean free paths:

ℓ_I : due to imperfections (residual bulk resistivity)

ℓ_T : due to phonons (ideal bulk resistivity)

The simple surface scattering (residual surface effects) can be approximated roughly by a simple argument as follows[14]:

Considering an electron starting at the center of the wire at an angle θ with the wire axis (see Figure III-1), we obtain a mean free path ℓ_{SS} by simply averaging over

all directions[+)]

$$\frac{1}{\ell_{ss}} = \frac{\pi}{4r} \quad \text{or} \quad \ell_{ss} = \alpha r \; ; \; \alpha : \text{constant}$$

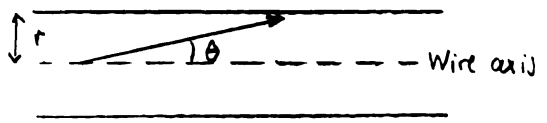

Figure III-1

It may be noted here that the result of our crude picture gives an answer for residual effects

$$\frac{1}{\ell_{resid.}} = \frac{1}{\ell_I} + \frac{1}{\ell_{ss}} = \frac{1}{\ell_I} + \alpha' \left(\frac{1}{d}\right)$$

- (α' : constant; d : diameter) - which agrees essentially with Dingle's rigorous result

$$\rho_{resid.} = \rho_{resid.}^{bulk} + \beta \left[\rho_{resid.}^{bulk} \ell_I / d\right]$$

(β : constant).

We now wish to find a mean free path ℓ_{TS} for the resistivity arising from electrons being brought to the surface by phonon interactions. It should be remarked here that the separation of the mechanisms listed under 2) and 4) above (p. 15) is here assumed possible, even though these processes are really not statistically independent.

+): For the objections to this procedure in a rigorous treatment, see Fuchs[3)]. We have essentially averaged over all paths of one electron rather than over all electrons at a given moment.

We now calculate the desired ℓ_{TS} as follows. At low temperatures each individual electron-phonon interaction produces scattering through an extremely small angle Θ [16]:

$$\Theta \leq \Theta_{max} \approx T/\Theta_D$$

where Θ_D is the Debye temperature. For larger angles the transition probability rapidly becomes vanishingly small. Thus many small angle scattering events will be necessary to bring an electron to the surface if it is initially moving along the wire axis. The average distance travelled before each collision (inversely proportional to the transition probability) is given by[17] (see Appendix, II.)

$$a = \left[\frac{(KT)^4}{8ne^2(mv)^3c^4}\right]\frac{1}{\rho_T^b}$$

where c is the velocity of sound, n is the number of electrons per unit volume, and ρ_T^b is the thermal bulk resistivity. We shall now assume that this distance is a constant for all scattering angles from zero to Θ_{max}; the problem then requires finding the mean free path for an electron undergoing multiple scattering such that

1) each individual event causes a scattering through an angle $\theta < \theta_{max}$;
2) the average distance of flight (and hence also the inverse of the transition probability) between two events is taken to be a constant (a) over all angles from zero to Θ_{max}, and infinite for all larger angles;
3) the electron, after having undergone a large number

of such collisions, is brought to the surface, where its momentum is randomized.

Starting with this information, ℓ_{TS} can be obtained (in a not totally unambiguous fashion) by a method developed in cosmic ray theory[18]. Consider the wire axis to be the x-axis of a Cartesian coordinate system (see Figure III-2); the radius of the wire is r. Then the probability that

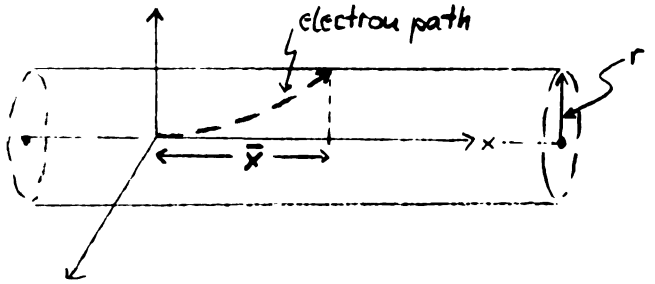

Figure III-2

an electron, initially moving along the wire axis, reaches the surface of the wire through multiple scattering after having traversed a distance $\bar{x}$ along the wire axis (cf. Figure III-2) is given by (see Appendix, II.)

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

where $$w = \left[2a/\pi\,\theta_{max}^4\right]^{1/2}$$

a being the "distance of flight" and θ_{max} the maximum scattering angle as defined above.

We now define as the mean free path ℓ_{TS} of this process that distance $\bar{x}_0$ along the wire axis, for which the probability that the electron has reached the surface is a

maximum. That is to say, we determine the distance $\bar{x}_0$ along the wire axis by maximizing $H(\bar{x})$ (the radius r is fixed). This procedure is ambiguous to the extent that the mean free path need not necessarily correspond to a maximum of $H(\bar{x})$ - one could equally well ask that the probability of the electron reaching the surface have any particular fixed value, such as, for instance, 1/e; furthermore, the mean free path is actually the path taken by the electron in going from the wire axis to a point on the surface --- not the corresponding distance along the wire axis. The first point, however, presumably does not intoduce any fundamentally significant difficulties+), whereas the second can be justified by assuming a sufficiently thin wire, so that the two distances are approximately equal. Finally we shall assume that the mean free path thus determined is approximately valid for all electrons (i.e., also those not initially moving along the wire axis). This then allows the most simple mathematical treatment.

Maximizing $H(\bar{x})$ with respect to $\bar{x}$ we find

$$\bar{x}_0 = \left[\omega^2 r^2\right]^{1/3} = \left[\frac{2a r^2}{\pi \theta_{max}^4}\right]^{1/3} = \left[\frac{m v}{4\pi n e^2}\right]^{1/3} \frac{r^{2/3}}{(\rho_T^b)^{1/3}}$$

where ρ_T^b was the thermal ("ideal") bulk resistivity and r the wire radius. We thus have as the approximate mean free path for thermal surface scattering

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+): Applying this procedure to thermal bulk scattering does result in essentially the correct ideal resistivity. See Appendix,III., where this consistency is shown.

$$\ell_{TS} = \left[\frac{mv}{4\pi ne^2}\right]^{1/3} \frac{r^{2/3}}{(\rho_T^b)^{1/3}}$$

Qualitatively we note that $1/\ell_{TS}$, and therefore the resistivity arising from this mechanism, is temperature dependent (through $(\rho_T^b)^{1/3}$), and that its contribution is greater for thinner wires, as one would expect and as found by Olsen.

Lastly we wish to consider electron-electron effects. The mean free path (in the bulk) for electron-electron collisions has been found by Abrahams[19] to be approximately

$$\ell_{ee} = \frac{1}{n\sigma_F}\left(\frac{E_F}{kT}\right)^2$$

where E_F is the Fermi energy and σ_F is the electron-electron scattering cross-section. σ_F is temperature independent since the Fermi energy remains quite constant over a wide temperature range. We note again that due to momentum conservation there will be no bulk resistive effects arising from electron-electron scattering.

We find an approximation to the mean free path for electron-electron-to-surface processes by a simple argument similar to the one used to find the simple surface scattering mean free path. Consider an electron moving along the wire axis; after a distance ℓ_{ee} it will experience an electron-electron collision in which the total electronic momentum is conserved; the mean distance S, which the electron now still has to traverse in order to reach the surface (where its momentum is randomized), was found before (for ℓ_{SS}) to be $4r/\pi$, r being the wire radius (see Figure III-3).

Hence here the total mean free path for the entire process in a thin wire is roughly

$$\ell_{se} = \ell_{ee} + \bar{s} = \ell_{ee} + \frac{4r}{\pi}$$

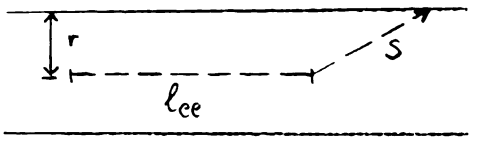

Figure III-3

We substitute in this the expression found by Abrahams for ℓ_{ee} and thus find

$$\ell_{se} = \frac{1}{n\sigma_F}\left(\frac{E_F}{KT}\right)^2 + \frac{4r}{\pi} = \frac{1}{(KT)^2}\left[\frac{E_F^2}{n\sigma_F} + \frac{4r}{\pi}(KT)^2\right]$$

Again we note qualitatively that $1/\ell_{se}$ and thus the resistivity due to this process increases as the wire radius is decreased; we also see that in the bulk ($r \longrightarrow \infty$) the effect vanishes, as expected, and finally, that the mechanism becomes more important as the temperature is increased.

Combining all our results, we find for the total mean free path in thin wires

$$\frac{1}{\ell_w} = \frac{1}{\ell_I} + \frac{1}{\ell_T} + \frac{1}{\ell_{ss}} + \frac{1}{\ell_{TS}} + \frac{1}{\ell_{se}} \qquad \text{(III-1)}$$

$$= \frac{1}{\ell_I} + \frac{1}{\ell_T} + \frac{\pi}{4r} + \left[\frac{4\pi ne^2}{m\upsilon}\right]^{1/3}\frac{(\rho_T^b)^{1/3}}{r^{2/3}} + \frac{(KT)^2}{E_F^2/n\sigma_F + \frac{4r}{\pi}(KT)^2}$$

Thus, for the thin wire resistivity, using the Sommerfeld theory expression

$$\rho = \left[\frac{m\upsilon}{ne^2}\right]\frac{1}{\ell}$$

we obtain the result

$$\rho_w = \rho_I^b + \rho_T^b + \left[\frac{\pi}{4}\rho_I^b \ell_I^b\right]\frac{1}{r} + (4\pi)^{1/3}\left[\frac{mv}{ne^2}\right]^{2/3}\frac{(\rho_T^b)^{1/3}}{r^{2/3}} \qquad \text{(III-2)}$$

$$+ \left[\frac{mv}{ne^2}\right]\left[\frac{(KT)^2}{E_F^2/n\sigma_F + 4r(KT)^2/\pi}\right]$$

where ρ_I^b and ρ_T^b are the bulk residual and ideal resistivities, respectively. This can be rewritten in a simpler form

$$\rho_w = \rho_I^b + \rho_T^b + A\left[\frac{\pi}{4r} + \left(\frac{4\pi}{A}\right)^{1/3}\frac{(\rho_T^b)^{1/3}}{r^{2/3}} + \frac{(KT)^2}{E_F^2/n\sigma_F + \frac{4r}{\pi}(KT)^2}\right] \qquad \text{(III-3)}$$

where $A = \left[\frac{mv}{ne^2}\right]$ is, for a given metal, a temperature and size independent constant.

IV. DISCUSSION AND COMPARISON WITH EXPERIMENTAL RESULTS

We had obtained the following expression for the resistivity of thin wires

$$\rho_w = \rho_I^b + \rho_T^b + A\left[\frac{\pi}{4r} + \left(\frac{4\pi}{A}\right)^{1/3}\frac{(\rho_T^b)^{1/3}}{r^{2/3}} + \frac{(K\bar{T})^2}{E_F^2/n\tau_F + \frac{4r}{\pi}(K\bar{T})^2}\right] \quad \text{(III-3)}$$

Briefly looking at the qualitative behaviour of (III-3) one sees that

1) for the bulk material, i.e., for $r \longrightarrow \infty$, each term within the square brackets vanishes and we obtain the correct bulk resistivity;

2) at $T = 0^\circ K$ all terms except ρ_I^b and $\frac{4r}{\pi}A$ vanish and we find the experimentally satisfactory Nordheim result

$$\rho_w(\bar{T}=0^\circ K) = \rho_I^b + \left(\frac{4A}{\pi}\right)\frac{1}{r}$$

3)the last two terms within the square brackets are temperature as well as size dependent, in contradiction to a strict matthiessen's rule, but in qualitative agreement with Olsen's results;

4) the size dependent part of the resistivity increases in importance the thinner the wires are made, and the total resistivity increases with <u>temperature</u> faster for thinner wires.

Hence we see that at least the qualitative behaviour of our result gives agreement with Olsen's experimental findings; we shall in the following attempt a somewhat more quantitative comparison.

A. Temperature Dependence

We shall consider here the temperature dependence of ρ_w without, for the moment, taking into account any electron-electron effects. Then we have

$$\rho_w(T) = \rho_w - \rho_w(0^\circ K) - \rho_{ee} = \rho_T^b + \left[\frac{(4\pi)^{1/3} A^{2/3} \rho_T^{b\,1/3}}{r^{2/3}} \right] \qquad \text{(IV-1)}$$

The value of A is determined from Olsen's experiments, by extrapolation of his measurements to $T = 0^\circ K$. We take for ρ_T^b the measured resistivity of the heaviest wires minus the extrapolated resistivity of these values for $T = 0^\circ K$. These wires exhibit approximately bulk behaviour.

In Figure IV-1[+)] both the calculated and the experimental values of $\rho_w(T)$ are plotted as a function of temperature. It is appearent that the calculated values are considerably too large and exhibit a slightly incorrect temperature dependence.

These discrepancies are possibly due to the following reasons:

1) We have considered only so-called "normal" electron-phonon scattering, which at low temperatures results in only small angle scattering of electrons. There exists, however, even at low temperatures, the possibility that the momentum of an electron is randomized through large angle scattering caused by direct interaction with the lattice

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+): Figures IV-1 to VI-6 are found at the end of Section IV.

("internal Bragg reflection"), i.e., via so-called "Umklapp"-processes. It has been calculated by Bailyn and Brooks[20] that in bulk sodium at 4°K Umklapp-processes give rise to about 80 % of the total resistivity, i.e., $\rho_u/\rho_N \approx 5$. It is almost certain that in the trivalent metal indium Umklapp-processes are at least as important as in sodium, the metal which most nearly approximates free electron gas behaviour. In all probability $\rho_u/\rho_N \gg 5$ for indium.

Since, however, in the derivation of the last term of (IV-1):

$$\left\{ (4\pi)^{1/3} A^{2/3} r^{-2/3} \right\} (\rho_T^b)^{1/3} \qquad \text{(IV-2)}$$

the bulk resistivity was assumed to be due to normal (small angle) processes only, the use of the total experimental bulk resistivity in the evalutation of (IV-2) must lead to a considerable overestimate of that term.

2) In the derivation of (III-3) the statistical independence of thermal bulk and thermal surface effects was assumed. This is undoubtedly not valid and may cause an error both in the values and in the temperature dependence of $\rho_w(\tau)$.

We now introduce a multiplicative parameter α in expression (IV-1) to account for these effects, i.e., we assume that

$$\rho_w(\tau) = \rho_b^\tau + \alpha \left\{ (4\pi)^{1/3} A^{2/3} r^{-2/3} \right\} (\rho_T^b)^{1/3} \qquad \text{(IV-3)}$$

and obtain α by matching experimental and theoretical curves at one temperature. We thus find the results shown (for a particular wire) in Figure IV-2. The general

comparison between experimental and theoretical temperature dependences is shown in Figure IV-3. These curves were obtained as follows: A plot of $\rho_W(T)$ vs. $r^{-2/3}$ should result in a series of straight lines (cf. Figure IV-4), each corresponding to a different temperature; the slopes of these lines should, according to our results, be proportional to $(\rho_T^b)^{1/3}$; their intercepts should be ρ_T^b . Comparing the experimental slopes (Figure IV-4) with $(\rho_T^b)^{1/3}$, we obtain Figure IV-3. It is seen that the temperature dependence of the calculated values in general is still somewhat too weak. This is also indicated by Figure IV-5; here a log-log plot of the slopes ($\sim \rho_{TS}$) vs. the intercepts (ρ_T^b) of Figure IV-4 is shown. According to our results we should obtain a straight line of slope 1/3 (cf. equation IV-2); the actual slope is somewhat steeper, indicating a somewhat stronger temperature dependence of the experimental values than that predicted by our model. A more quantitative statement does not seem justified here, since only a small number of somewhat randomly distributed experimental values are available in this case.

Of the above reasons for the quantitative failure of the theoretical results, the first could be eliminated, provided the effect of Umklapp-processes on the total bulk resistivity were known. The appropriate changes in the calculations could then be made. To our knowledge, however, there exist at the present time neither calculations nor any experimental data which might allow a reliable

estimate of ρ_u/ρ_N +). Such correction, when possible, would however not lead to any significant change in the temperature dependence of $\rho_w(T)$, since normal and Umklapp-processes show approximately the same temperature dependence[22].

It would seem difficult to correct for the second objection, i.e., the statistical correlation of thermal bulk and surface processes (which, as a consequence of the above remarks, would have to account for the temperature dependence discrepancies), within the frame-work of our simple model. A suitable parameter α, as used above, would, if it were made to include effects arising from the statistical correlation of the two types of thermal mechanisms, become both temperature and size dependent. The crudeness of our model, however, would hardly warrant or justify such elaborate extensions.

B. Size Dependence

At T = 0°K our results reduce to Nordheim's[14] expression; Olsen showed that his results extrapolated to T = 0°K agree quite well with this expression. At higher temperatures, according to equation (IV-1), one would expect the temperature dependent resistivity $\rho_w(T)$ to go as

$$\rho_w(T) = \rho_T^b + f(T)\, r^{-2/3} \qquad \text{(IV-4)}$$

+): One could, for example, estimate the ratio ρ_u/ρ_N from the ratio of electrical to thermal conductivity at low temperatures[21], if this information were available.

Hence a plot of the experimental values of $\rho_w(T)$ vs. $r^{-2/3}$ should result, according to our model, in a series of straight lines (each line corresponding to a particular temperature), intersecting the $\rho_w(T)$-axis at the value of the bulk thermal resistivity ρ_T^b for that temperature (cf. p. 26 also).

Such a plot is shown in Figure IV-4; it is seen that the general behaviour of the experimental results agrees with our predictions. However, not enough experimental values are available to reach very definite quantitative conclusions.

C. Electron-Electron Effects

The temperature variation of the electron-electron-to-surface effects alone can be compared with the temperature behaviour of the various other mechanisms. In Figure IV-6 a plot against temperature of the experimental values for the total thermal resistivity $\rho_w(T)$ and the bulk thermal resistivity ρ_T^b, as well as for the theoretical values for the surface mechanisms is shown. (The latter are matched with $\rho_w(T)$ at 3°K for comparison purposes). It would seem rather unlikely, however, that the electron-electron effect, as treated here, is responsible for anything but a fairly small fraction of the total thermal wire resistivity, since the latter does show quite a pronounced size dependence, whereas the electron-electron term, at least in our model, gives rise to a resistivity contribution which is quite insensitive to size variations. We are forced to this conclusion

by the fact that in our result

$$\rho_{se} = A \Big/ \left[\frac{E_F^2}{n\sigma_F} \left(\frac{1}{K\bar{r}}\right)^2 + \frac{4r}{\pi} \right] \qquad \text{(IV-5)}$$

the first term in the denominator (treating $E_F^2/n\sigma_F$ as a parameter) has to be considerably larger than the second in order to obtain a quantitatively reasonable contribution form this effect.

D. Comparison of Various Mechanisms

We see that our model provides a qualitatively reasonable picture of lattice scattering of electrons and the associated surface effects in thin wires; this picture can be improved quantitatively by considering Umklapp-processes. Electron-electron effects, in our approach, seem to give rise to only a lesser contribution, which is quite insensitive to size variations.

It is appearent that we have made numerous simplifying approximations in the development of our model --- approximations which are not merely difficult to justify, but which may very possibly lead to serious errors. In support of our crude model we can only cite the success of Nordheim's equally crude approach. --- Quite aside from the above considerations, a number of approximations are implicit in our treatment as well as in all previous work, such as that of Fuchs and Dingle.

First we have not considered the effect that localized surface states (Tamm states) might have on the conductivity of thin wires. Practically nothing is known of surface

states in metals, although during recent years investigations of this field have been initiated[23].

Second, we have assumed throughout, as did all other workers, that a continuum of states in momentum space is available to electrons and phonons. Now it is clear that in a thin wire the number of vibrational modes with wave vectors normal to the wire axis is $3\ N_a$, where N_a is the number of atoms in a cross-sectional area of the wire. For thin wires N_a may be sufficiently small so that at low temperatures the energy difference between neighbouring vibrational modes is of order kT. In that case the usual description (density of states, etc.) for normal electron-phonon scattering must fail[24]. Size effects attributable to the discreteness of the phonon spectrum have been observed in the past, most recently by Tanttila and Jennings[25]. The same boundary conditions which lead to a discrete (not even quasi-continuous) phonon spectrum will similarly also give rise to a discrete electron spectrum, as distinguished from the quasi-continuous spectrum for the conduction electrons in the bulk metal. Hence a treatment which assumes a continuous spectrum cannot be correct.[26]

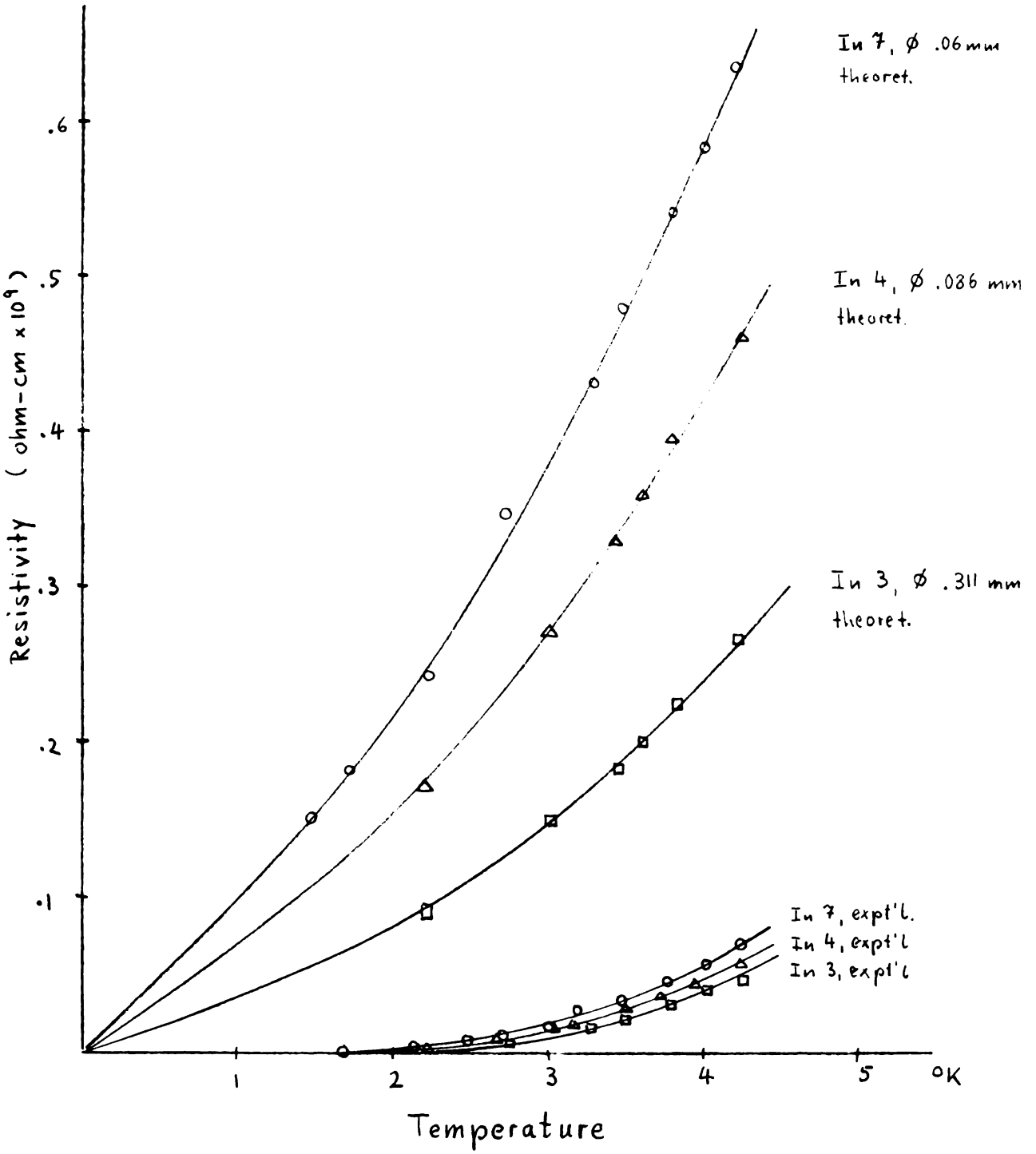

Figure IV-1

Experimental and theoretical values for the temperature variation of thin wire resistivity (unadjusted)

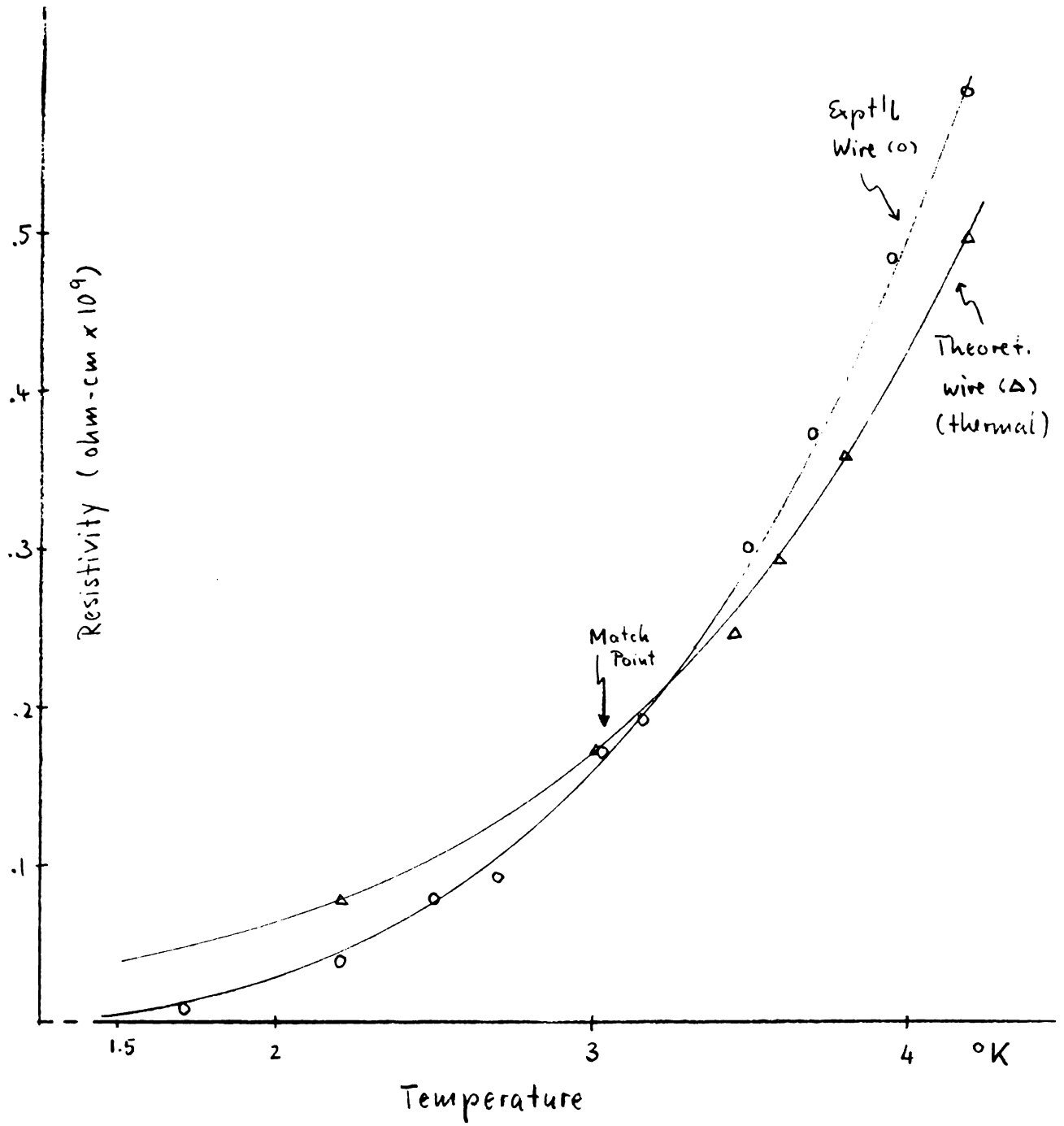

Figure IV-2

Experimental and theoretical (thermal only) temperature variation of thin wire resistivity for a particular wire (In 4); matched at 3°K.

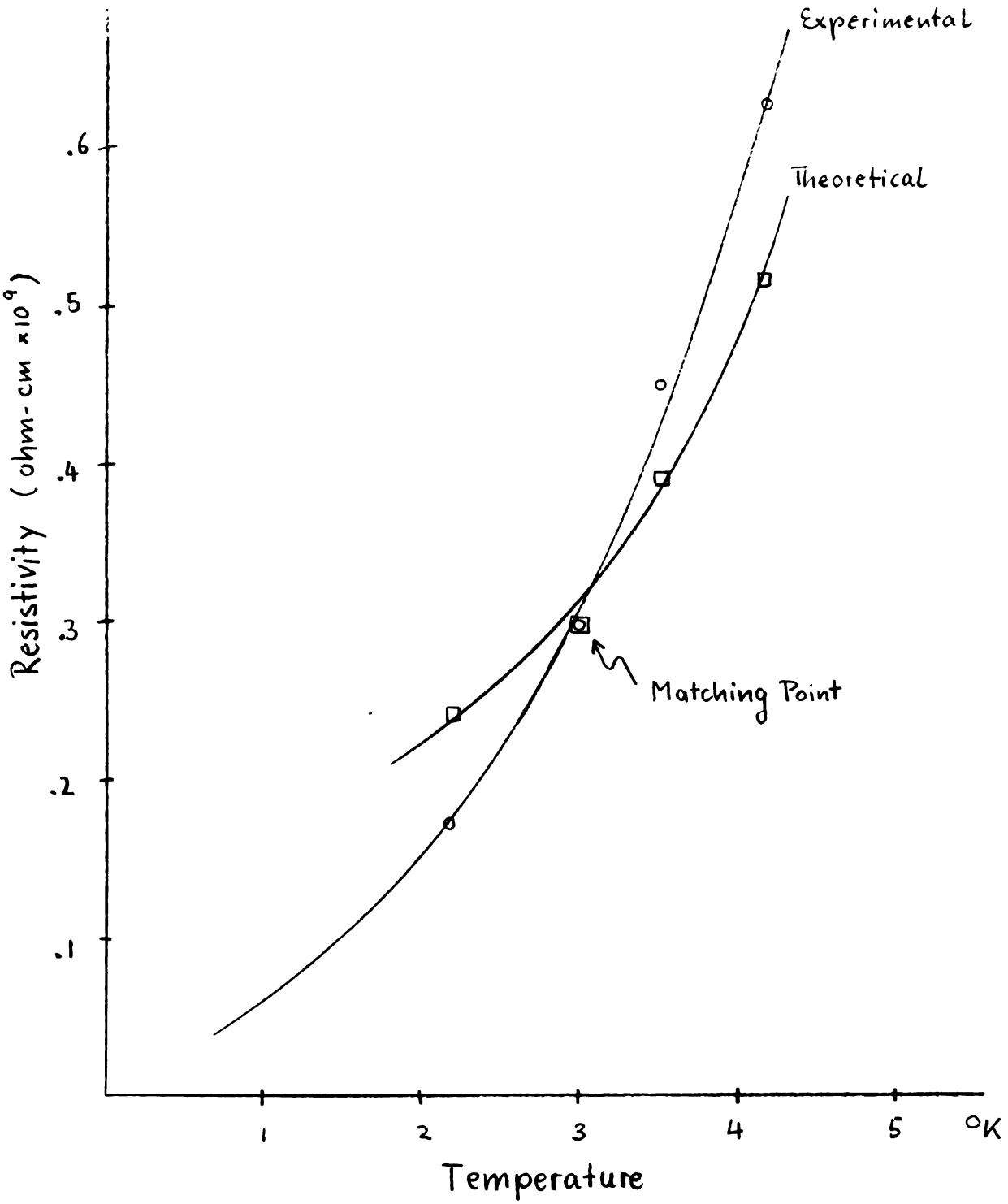

Figure IV-3

Comparison of general temperature dependence, experimental and theoretical, of thin wire resistivity (matched at 3°K)

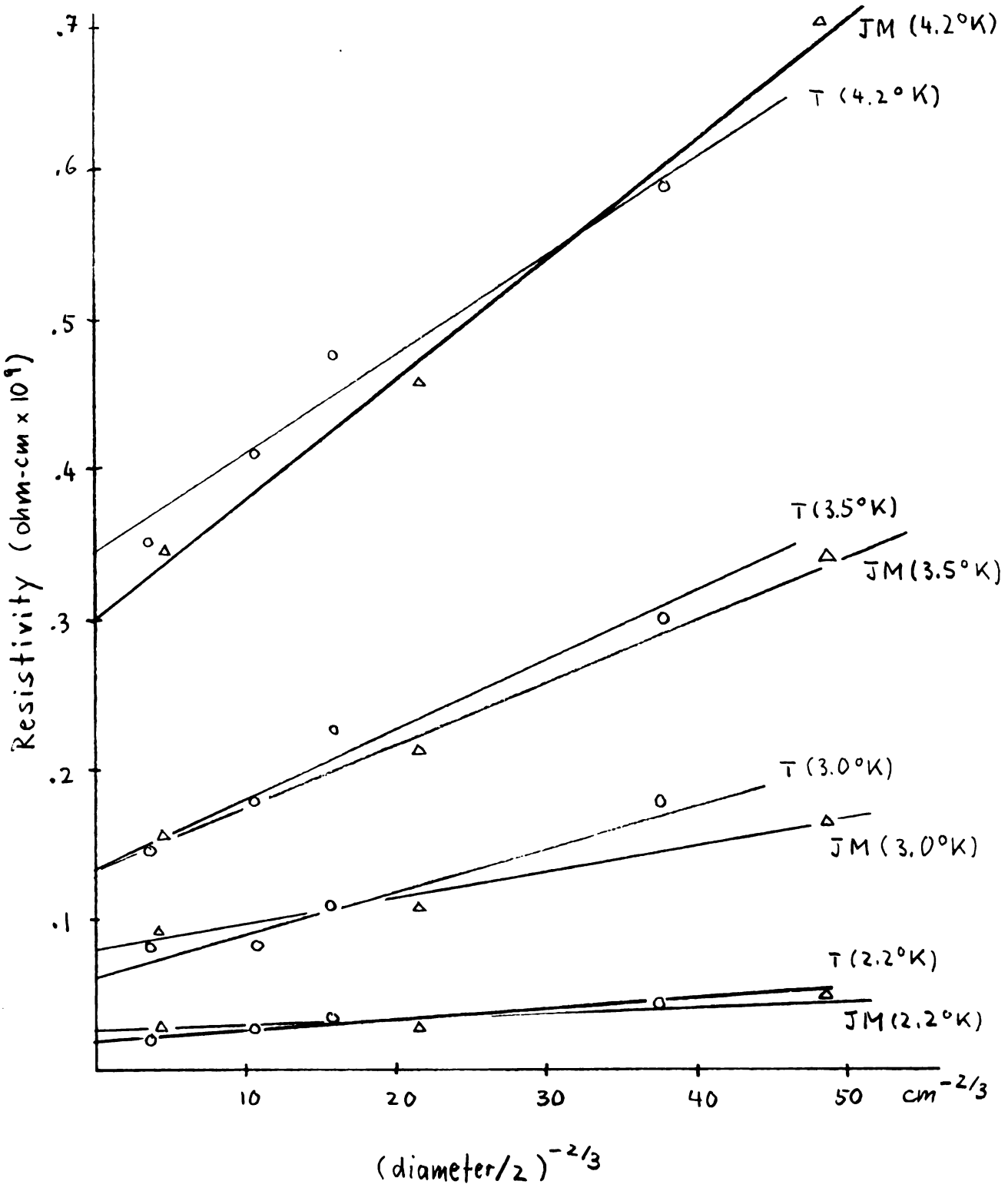

Figure IV-4

Experimental size dependence of thin wire resistivity, at various temperatures.

(o): Tadanac Brand (T) ; (Δ): Johnson, Matthey (J M)

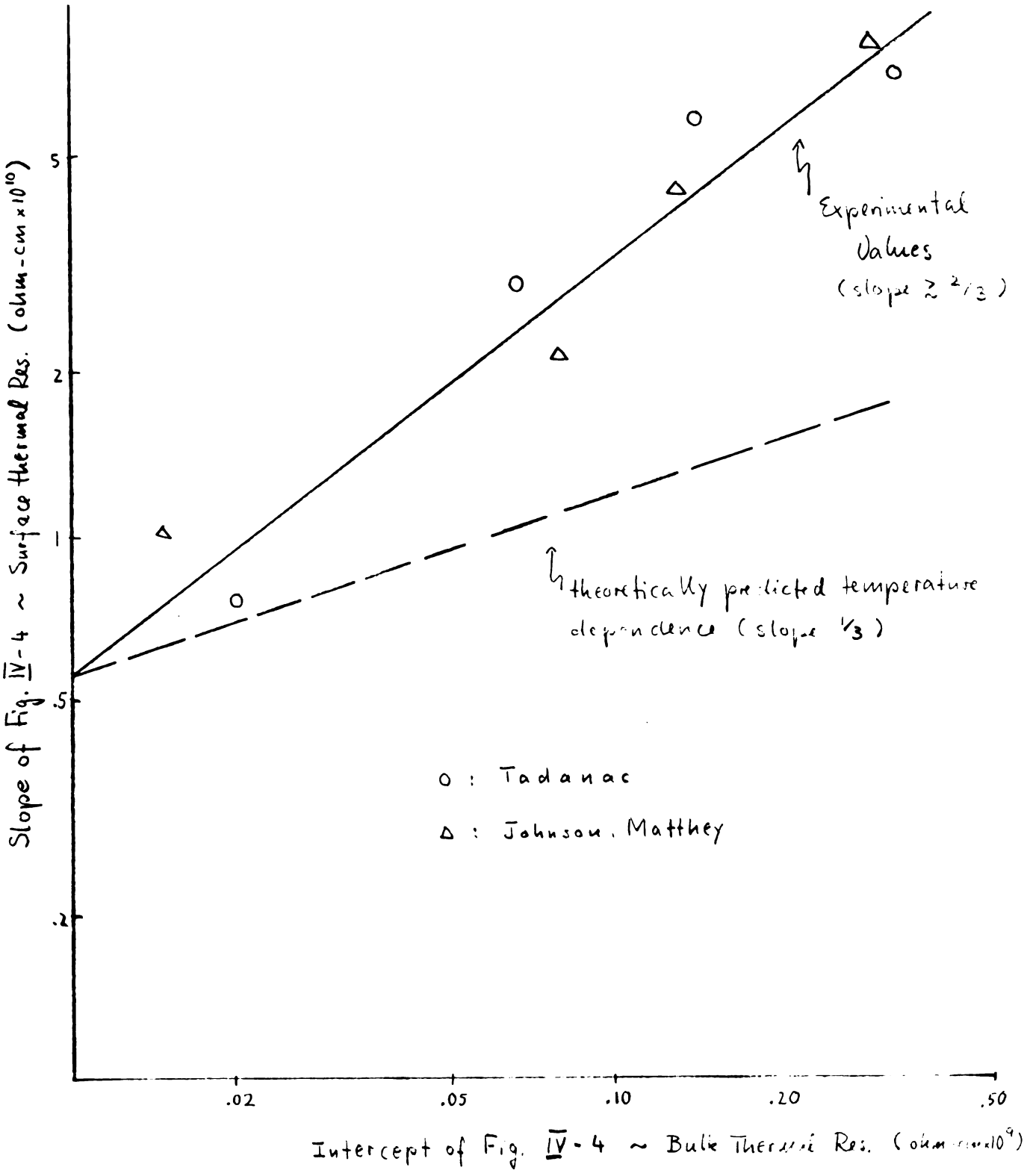

Figure IV-5

Experimental temperature dependence of thin wire resistivity (cf. p. 26 for further explanation)

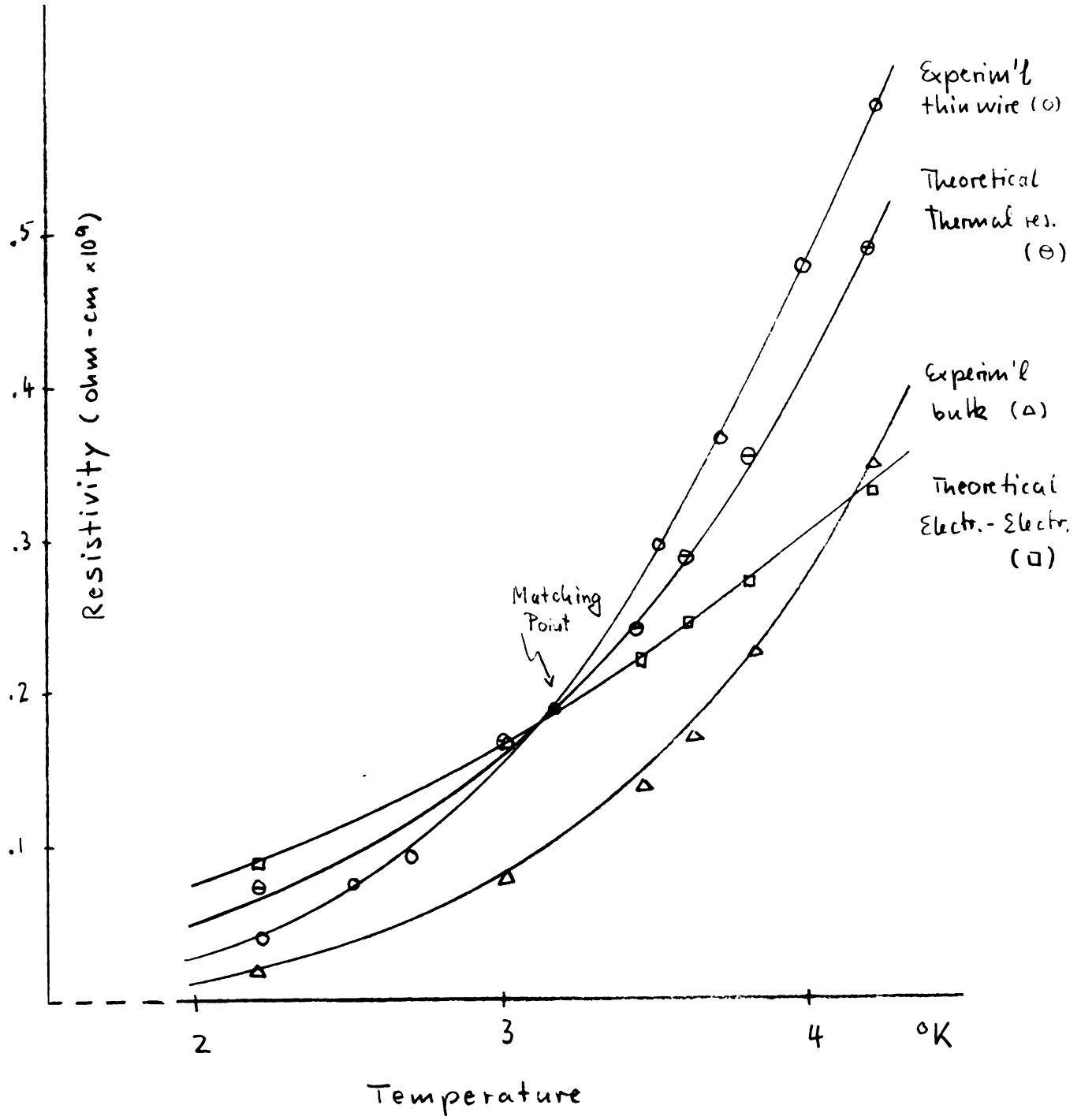

Figure IV-6

Comparison of experimental wire and bulk resistivity with various theoretical resistivity effects (matched at 3°K, except for bulk values) for a particular wire

(In 4)

V. CONCLUSION

The general nature of our results indicates, as should perhaps be expected, that the simple model used to investigate the various mechanisms giving rise to resistivity in thin wires provides a picture which shows qualitatively fair agreement with experimental results, but which is insufficient to give correct or even conclusive quantitative answers. Many difficulties arising in a Fuchs type treatment have been avoided only by using the crudest physical and mathematical approach possible; any great elaboration would re-introduce these same problems also in this type of kinetic approach. It has been shown that a mechanism such as suggested by Olsen can account, at least to some extent, for the temperature dependence of the resistivity of thin wires, and, further, that effects due to Umklapp-processes and electron-electron-to-surface mechanisms should at any rate not be excluded a priori in a more rigorous future investigation. Before the relative importance of these mechanisms can be evaluated, however, further investigations on bulk phenomena (such as a determination of the contribution of Umklapp-processes to the total bulk resistivity, or the calculation of electron-electron mean free paths in bulk materials) would have to be undertaken.

Even though it would seem that in the samples investigated by Olsen electron-electron scattering is relatively unimportant, the arguments concerning the relative importance of such effects in thin wires and in the bulk are

presumably correct. It follows that suitable measurements in thin wires may provide information on a mechanism, which, although it exists, cannot be observed in bulk materials.

VI. APPENDIX

I. Derivation of the "distance of flight" for electron-phonon scattering.

If $a(\theta)$ is the "distance", $\bar{\tau}(\theta)$ the "time" of flight between two collisions, then

$$a(\theta) = v\,\bar{\tau}(\theta)$$

where θ is the scattering angle.

Now

$$\rho_b^{\bar{\tau}} = \frac{m}{ne^2}\,\frac{1}{\bar{\tau}} = \frac{m}{ne^2}\int \frac{1}{\bar{\tau}(\theta)}\,(1-\cos\theta)\sin\theta\,d\theta$$

and if we assume $\bar{\tau}(\theta) = \text{constant} = \tau'$ for $0 \le \theta \le \theta_{max}$

$$= \infty \quad \text{for} \quad \theta > \theta_{max}$$

and $\theta_{max} \ll 1$

then

$$\rho_T^b \approx \frac{m}{ne^2}\,\frac{1}{\tau'}\int_0^{\theta_{max}} \theta_{/2}^3\,d\theta = \frac{m}{ne^2}\cdot\frac{1}{\tau'}\cdot\frac{\theta_{max}^4}{8}$$

Hence

$$\tau' = \left[m\,\theta_{max}^4\right] \Big/ \left[8ne^2\rho_b^{\bar{\tau}}\right]$$

But we have[17]

$$\theta_{max} = \frac{KT}{mvc}$$

and thus obtain

$$a = v\,\bar{\tau}' = \left\{\frac{(KT)^4}{[8ne^2(mv)^3c^4}\right\}\frac{1}{\rho_T^b}$$

which is the cited result.

II. Outline of the multiple scattering probability derivation.

(1) Single Event.

Here we know the probability per unit time that an electron is scattered, by a single collision, through an angle Θ into a solid angle $d\omega$:

$$P_t(\theta)\, d\omega \quad \text{where} \quad P_t(\theta) = v/a \quad \text{and} \quad \left\{ \begin{array}{l} a : \text{distance of flight} \\ 0 \le \theta \le \theta_{max} \end{array} \right\}$$

Then $P_t(\theta)\, d\omega = v\, P_\ell(\theta)\, d\omega$

where $P_\ell(\theta)$ is the corresponding probability per unit <u>length</u> traversed. Hence

$$P_\ell(\theta) = \frac{1}{v} P_t(\theta) = \frac{1}{a}, \quad 0 \le \theta \le \Theta_{max}$$

Then the mean square scattered angle for an electron which has traversed a small distance Δx is

$$\langle \theta^2 \rangle_{\Delta x} = \Delta x \int_0^{\theta_{max}} \theta^2 P_\ell(\theta)\, \theta\, d\theta \cdot 2\pi = \frac{1}{w^2} \Delta x$$

where

$$w = \left[2a/\pi\, \theta_{max}^4 \right]^{1/2}$$

(2) Multiple Scattering.

We cannot use here immediately $P_\ell(\theta)$, which was the transition probability for the <u>single</u> event. Instead we use a multiple scattering transition probability $\rho_{\Delta x}(\theta)$ defined such that

$$\int_{-\infty}^{\infty} \rho_{\Delta x}(\theta)\, d\theta = 1$$

and $$\int_{-\infty}^{\infty} \theta^2 \rho_{\Delta x}(\theta)\, d\theta = \langle \theta^2 \rangle_{\Delta x} = \frac{1}{\omega^2}\, \Delta x$$

where the limits are extended since $\rho_{\Delta x}(\theta)$ is assumed to have a sharp maximum about $\theta = 0$ and then go to zero. The $\rho_{\Delta x}(\theta)$ thus defined gives the probability of an electron being scattered through an angle θ by <u>many</u> scatterings (each of which is through a small angle) after having traversed a distance Δx along the direction of initial motion (the x-axis). If the angle of "multiple" scattering or the traversed distance Δx is made very small, the problem should reduce to the single scattering event, hence the second of the above requirements.

Using this probability, one finds[18] the probability $G(\bar{x}, \theta)$ that an electron, after traversing a distance $\bar{x}$, is deflected by an angle θ from its initial direction, and further the probability $H(\bar{x}, s)$ that an electron, again after going a distance $\bar{x}$ along the wire axis, is deflected a distance s perpendicular to the wire axis. These two probabilities are given by

$$G(\bar{x}, \theta) = \frac{\omega}{2\sqrt{\pi}(\bar{x})^{1/2}} \exp\left[-\tfrac{1}{4}\omega^2\theta^2(\bar{x})^{-1}\right]$$

$$H(\bar{x}, s) = \frac{\omega\sqrt{3}}{2\sqrt{\pi}(\bar{x})^{3/2}} \exp\left[-\tfrac{3}{4}\omega^2 s^2(\bar{x})^{-3}\right].$$

III. Consistency check: Bulk thermal resistivity via multiple scattering method.

In the bulk material, the electron momentum is randomized if the electron has undergone enough scatterings to be deflected by $\pi/2$ from the original direction of motion. Hence we maximize $G(\bar{x}, \theta)$ for $\theta = \pi/2$ and find

$$\overline{x_0} = \frac{1}{8} w^2 \pi^2$$

Thus

$$\bar{\tau} = \frac{1}{v} (w^2 \pi^2/8)$$

and

$$\rho_T^b = \frac{m}{ne^2} \frac{1}{\bar{\tau}} = \frac{mv}{ne^2} \frac{8}{\pi^2} \frac{4\pi ne^2}{mv} \rho_T^b$$

$$= \frac{32}{\pi^2} \rho_T^b$$

Hence our result obtained in this fashion is wrong by a constant multiplicative factor of $32/\pi^2$ only, giving the "correct" answer otherwise.

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