JOURNAL OF MATERIALS SCIENCE 21 (1986) 1405–1412

Numerical approximations for transport parameters in the framework of multidimensional conduction models

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General expressions for the electrical conductivity, the temperature coefficient of resistivity, the thermoelectric power, the thermal conductivity and the Hall coefficient of thin metal films are proposed. The basic functions are extensively tabulated, and linearized general expressions are obtained in this way; they are in agreement with approximate equations obtained previously. Physical consequences are deduced in good agreement with previous data.

1. Introduction

Recent studies [1, 2] related to the electrical resistivity of thin metal films, the temperature coefficient of resistivity and the Hall coefficient at low magnitude field lead to linearized expressions [1, 3, 4] which exhibit similar forms.

The aim of this paper is to give general expressions for the electrical conductivity, its temperature coefficient and the Hall coefficient; to tabulate their values; and to propose approximate linearized expressions, whatever the film structure, i.e. monocrystalline, columnar or polycrystalline.

2. Theoretical models

From theoretical studies in the framework of three-dimensional [1, 5] or bidimensional [1, 2, 4, 6] models of conduction the following general formulations for the electrical resistivity ϱ_f or conductivity σ_f, its temperature coefficient (t.c.r.) β_f, and the Hall coefficient (at low magnetic field) of thin metal films can be proposed, whatever the film structure:

$$\sigma_f/\sigma_o = A(b, \gamma) \quad (1)$$

$$\beta_f/\beta_o = B(b, \gamma)\,[A(b, \gamma)]^{-1} \quad (2)$$

$$R_{Hf}/R_{Ho} = B(b, \gamma)\,[A(b, \gamma)]^{-2} \quad (3)$$

with

$$b = \mu^{-1} + C_1 \nu^{-1} \quad (4)$$

$$\gamma = b^{-1}(1 + C^2 \nu^{-1}) \quad (5)$$

$$\mu = a\lambda_o^{-1}\left(\ln\frac{1}{p}\right)^{-1} \qquad p > 0.3 \quad (6')$$

$$\nu = D_g\lambda_o^{-1}\left(\ln\frac{1}{t}\right)^{-1} \qquad t > 0.3 \quad (7')$$

$$\mu = a\lambda_o^{-1}(1 + p)\,[2(1 - p)]^{-1} \quad (6'')$$

$$\nu = D_g\lambda_o^{-1}(1 + t)\,[2(1 - t)]^{-1} \quad (7'')$$

without any restriction in p and t;

$$A(b, \gamma) = \frac{3}{2b}\left[\gamma - \tfrac{1}{2} + (1 - \gamma^2)\ln(1 + \gamma^{-1})\right] \quad (8)$$

$$B(b, \gamma) = \frac{3}{2b^2}\left[\gamma^{-1} - 2 + 2\gamma\ln(1 + \gamma^{-1})\right] \quad (9)$$

where C_1 is a constant depending on the structure of the film ($C_1 = 1 - C$ in the case of polycrystalline films [1]; $C_1 = -C$ in the case of monocrystalline [6] and columnar [2] films). C is a constant equal to $4/\pi$, a is the film thickness, λ_o the electron mean free path in the bulk material, p the usual [1, 7] specular reflection coefficient at the film surface, D_g the grain diameter, t the statistical transmission coefficient at a grain boundary [1, 8] and the subscript o refers to the bulk material.

Let us remember that the thermoelectric power, S_f, can also be expressed in terms of the reduced t.c.r. [9, 10], provided that the only dependence on electron energy in the size effect and grain-boundary function is due to that in λ_o; it becomes

$$S_f = -\frac{\pi^2 B^2 T}{3eE_F}\left[U + V\frac{\beta_f}{\beta_o}\right] \quad (10)$$

where B is the Boltzmann constant, T the absolute temperature, $-e$ the electronic charge, E_F the Fermi energy, and

$$U = \left.\frac{\partial \ln \sigma_o}{\partial \ln E}\right|_{E=E_F} \quad (11)$$

$$V = \left.\frac{\partial \ln \lambda_o}{\partial \ln E}\right|_{E=E_F} \quad (12)$$

where E is the electron energy.

Moreover, in the case of noble metals the thermal conductivity due to electrons, C_f, is deduced from the electrical conductivity according to the Wiedemann-Franz law [10, 11] which states that

$$\frac{C_f}{\sigma_f T} = L \quad (13)$$

where L is the Lorentz constant, given by

$$L = \pi^2 B^2/3e^2 \quad (14)$$

In the case of polycrystalline structure Equation 4

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becomes [1]

$$b = b_3 = \mu^{-1} + (1 - C)v^{-1} \quad (15)$$

In the case of columnar or monocrystalline films, b becomes [1, 2]

$$b = b_2 = \mu^{-1} - Cv^{-1} \quad (16)$$

In the case where no grain-boundary scattering is operative, b reduces to

$$b = b_1 = \mu^{-1} \quad (17)$$

Equations 15 to 17 and Equations 1, 2, 3, 10 and 13 show that Equations 8 and 9 allow a complete quantitative description of the transport properties of metal films.

3. Numerical results

3.1. Tabulated values of $A(b, \gamma)$ and $B(b, \gamma)$

Numerical values of $A(b, \gamma)$ and $B(b, \gamma)$ have been calculated (Table I) with the aid of a microcomputer, limiting our attention to the values of b and γ satisfying the relation

$$|b\gamma| > 1 \quad (18)$$

which is a consequence of Equations 4 and 5.

TABLE I Numerical values of $A(1, \gamma)$, $B(1, \gamma)$ and $B(1, \gamma)/A^2(1, \gamma)$

γ	$A(1, \gamma)$	$B(1, \gamma)$	$B(1, \gamma)/A^2(1, \gamma)$
0.050	3.8803	27.4566	1.8234
0.100	2.9608	12.7193	1.4508
0.150	2.4615	7.9165	1.3065
0.200	2.1301	5.5750	1.2286
0.250	1.8882	4.2070	1.1799
0.300	1.7015	3.3197	1.1465
0.350	1.5518	2.7031	1.1224
0.400	1.4284	2.2533	1.1042
0.450	1.3246	1.9129	1.0900
0.500	1.2359	1.6479	1.0788
0.550	1.1590	1.4368	1.0696
0.600	1.0915	1.2654	1.0620
0.650	1.0319	1.1242	1.0556
0.700	0.9787	1.0061	1.0502
0.750	0.9310	0.9064	1.0456
0.800	0.8879	0.8212	1.0416
0.850	0.8487	0.7478	1.0382
0.900	0.8129	0.6841	1.0351
0.950	0.7801	0.6284	1.0324
1.00	0.7500	0.5794	1.0301
1.05	0.7221	0.5360	1.0279
1.10	0.6963	0.4975	1.0261
1.15	0.6723	0.4630	1.0243
1.20	0.6499	0.4320	1.0228
1.25	0.6290	0.4041	1.0214
1.30	0.6094	0.3789	1.0201
1.35	0.5911	0.3560	1.0190
1.40	0.5738	0.3352	1.0179
1.45	0.5575	0.3161	1.0169
1.50	0.5422	0.2987	1.0160
1.60	0.5139	0.2679	1.0145
1.70	0.4884	0.2417	1.0131
1.80	0.4654	0.2192	1.0119
1.90	0.4445	0.1997	1.0109
2.00	0.4254	0.1827	1.0100
2.10	0.4078	0.1679	1.0092
2.20	0.3917	0.1547	1.0085
2.30	0.3768	0.1431	1.0079
2.40	0.3630	0.1328	1.0073
2.50	0.3502	0.1235	1.0068
3.00	0.2978	0.0891	1.0050
4.00	0.2292	0.0527	1.0030
5.00	0.1864	0.0348	1.0019
6.00	0.1570	0.0247	1.0014
7.00	0.1357	0.0184	1.0010
8.00	0.1195	0.0142	1.0008
9.00	0.1067	0.0114	1.0006
10.00	0.0964	0.0093	1.0005
11.00	0.0879	0.0077	1.0004
12.00	0.0808	0.0065	1.0003
13.00	0.0747	0.0055	1.0003
14.00	0.0695	0.0048	1.0002
15.00	0.0650	0.0042	1.0002
16.00	0.0610	0.0037	1.0002
17.00	0.0575	0.0033	1.0002
18.00	0.0544	0.0029	1.0001
19.00	0.0516	0.0026	1.0001
20.00	0.0490	0.0024	1.0001
21.00	0.0467	0.0021	1.0001
22.00	0.0446	0.0019	1.0001
23.00	0.0427	0.0018	1.0001
24.00	0.0410	0.0016	1.0001
25.00	0.0394	0.0015	1.0000
26.00	0.0379	0.0014	1.0000
− 1.25	1.2670	1.8353	1.1432
− 1.30	1.1823	1.5648	1.1194
− 1.35	1.1095	1.3560	1.1015
− 1.40	1.0460	1.1901	1.0877
− 1.45	0.9899	1.0553	1.0767
− 1.50	0.9401	0.9437	1.0678
− 1.55	0.8953	0.8500	1.0604
− 1.60	0.8548	0.7704	1.0543
− 1.65	0.8180	0.7021	1.0490
− 1.70	0.7844	0.6428	1.0446
− 1.75	0.7536	0.5911	1.0407
− 1.85	0.6990	0.5054	1.0344
− 1.95	0.6519	0.4376	1.0295
− 2.05	0.6110	0.3829	1.0256
− 2.15	0.5750	0.3381	1.0224
− 2.25	0.5431	0.3008	1.0198
− 2.35	0.5146	0.2695	1.0176
− 2.45	0.4890	0.2430	1.0158
− 2.55	0.4659	0.2202	1.0142
− 2.65	0.4449	0.2005	1.0129
− 2.75	0.4257	0.1834	1.0118
− 2.85	0.4081	0.1684	1.0108
− 3.00	0.3844	0.1491	1.0095
− 4.00	0.2771	0.0771	1.0048
− 5.00	0.2168	0.0471	1.0029
− 5.80	0.1847	0.0341	1.0021
− 6.00	0.1781	0.0317	1.0019
− 7.00	0.1511	0.0228	1.0013
− 8.00	0.1312	0.0172	1.0010
− 9.00	0.1160	0.0134	1.0008
−10.00	0.1039	0.0108	1.0006
−12.00	0.0860	0.0074	1.0004
−14.00	0.0734	0.0053	1.0003
−16.00	0.0640	0.0040	1.0002
−18.00	0.0567	0.0032	1.0001
−20.00	0.0509	0.0025	1.0001
−22.00	0.0462	0.0021	1.0001
−24.00	0.0423	0.0017	1.0000
−26.00	0.0390	0.0015	1.0000
−28.00	0.0362	0.0013	1.0000
−30.00	0.0337	0.0011	1.0000
−32.00	0.0316	0.0010	1.0000
−34.00	0.0297	0.0008	1.0000
−36.00	0.0280	0.0007	1.0000
−38.00	0.0265	0.0007	1.0000
−40.00	0.0252	0.0006	0.9999
−42.00	0.0240	0.0005	0.9999
−44.00	0.0229	0.0005	1.0000
−46.00	0.0219	0.0004	0.9998
−48.00	0.0209	0.0004	1.0000
−50.00	0.0201	0.0004	0.9998

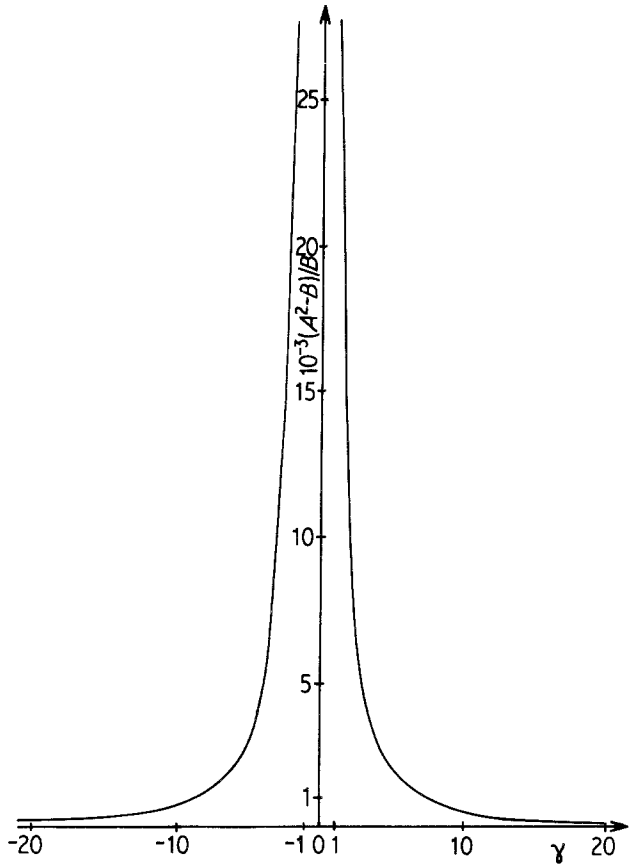

Figure 1 Relative deviation of A^2 from B (Equations 8 and 9) with γ (Equation 5); the value of b (Equation 4) is irrelevant in the range extending from -10 to $+100$.

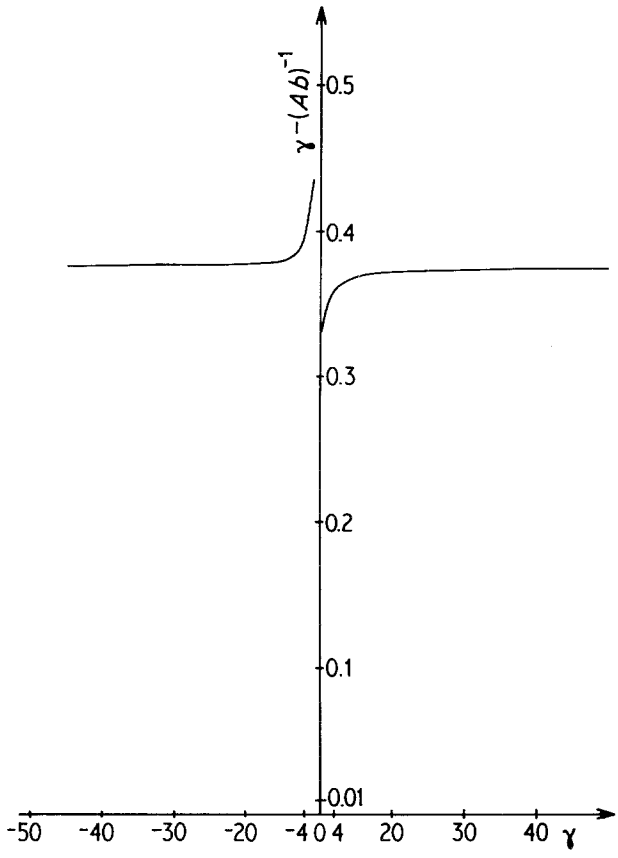

Figure 2 Variation in γ-$(Ab)^{-1}$ with γ. The value of b is irrelevant ($-10 < b < 100$).

Since C_1 takes negative value, Equation 4 shows that b can take negative values; the minimal value b_{min} is obtained for $1/\mu = 0$ so that

$$b_{min} = C_1/\nu$$

The corresponding value of γ is

$$\gamma_{min} = \frac{\nu + C^2}{C_1} \leqslant \gamma_m < 0$$

with

$$\gamma_m = \frac{C^2}{C_1}$$

where

$$\gamma_m = \frac{C^2}{1 - C} \approx -6 \qquad \text{for polycrystalline films}$$

$$\gamma_m = -C \approx -1.28 \qquad \text{for monocrystalline and columnar films}$$

Hence the range of values for γ in Table I is $\gamma < \gamma_m < 0$ and $\gamma > 0$.

Since it has been theoretically predicted [12, 13] that the Hall coefficient at low magnetic field exhibits a slight size effect, the values of A^2 have been computed (Table I). It appears that the relation

$$A^2 \approx B \qquad (19)$$

is valid in a large domain of the values of b and γ (Fig. 1).

Numerical values of the quantity $-[\gamma - (Ab)^{-1}]$ have been computed showing that the relation

$$\gamma - (Ab)^{-1} \approx -0.375 \qquad (20)$$

is valid in a large domain (Fig. 2). Consequently an approximate linearized expression for A is

$$A \approx [b\gamma + C_2 b]^{-1} \qquad (21')$$

with $C_2 = 0.375$. An alternative form is

$$A \approx [1 + (C^2 + C_1 C_2)\nu^{-1} + C_2\mu^{-1}]^{-1}$$
$$p, t > 0.3 \qquad (21'')$$

Tabulated values of A have been computed from Equations 8 and 21′ (Fig. 3).

When Equations 8 and 9 are expanded in power of γ^{-1} the following equations are obtained:

$$A(b, \gamma) \approx \frac{3}{2b} \ln \gamma^{-1} - \frac{3}{4b} \qquad (22)$$

$$B(b, \gamma) \approx \frac{3}{2b^2\gamma} - \frac{3}{b^2} \qquad (23)$$

When Equation 21′ is not valid (Fig. 2) the above equations hold (Figs. 4 and 5).

3.2. Approximate expressions for the electrical parameters at large thickness

Starting from Equations 1, 2, 3, 10 and 13 the following equations are valid in the validity range of Equation 21″:

$$\frac{\sigma_o}{\sigma_f} = \frac{\varrho_f}{\varrho_o} \approx 1 + \frac{C^2 + C_1 C_2}{\nu} + \frac{C_2}{\mu} \qquad (24)$$

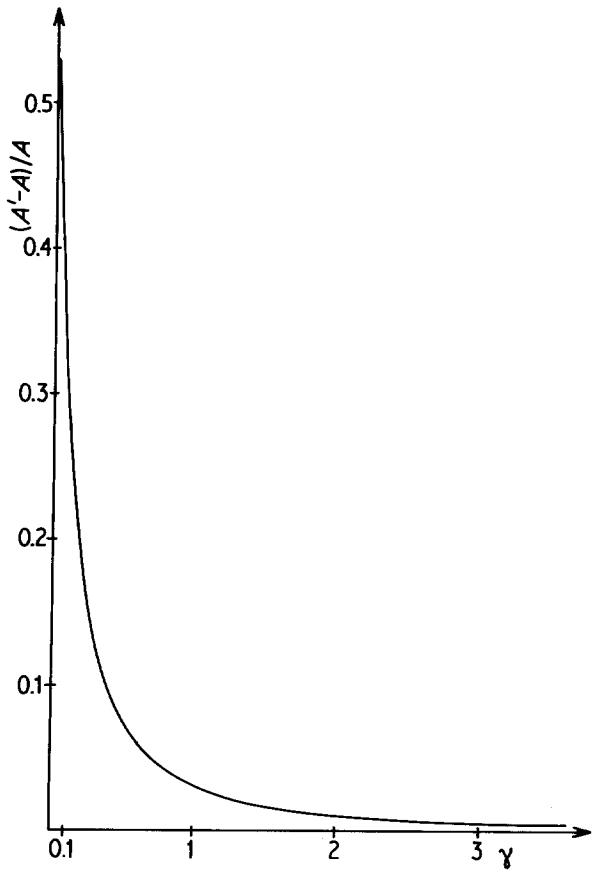

Figure 3 Relative deviation of A' (Equation 21′) from A (Equation 8) with γ (Equation 5). The value of b is irrelevant ($-10 < b < 100$).

$$\beta_f/\beta_o \approx \varrho_o/\varrho_f \tag{25}$$

$$R_{Hf}/R_{Ho} \approx 1 \tag{26}$$

$$S_f = -\frac{\pi^2 B^2 T}{3eE_F}\left[U + V\frac{\beta_f}{\beta_o}\right] \approx -\frac{\pi^2 B^2 T}{3eE_F}\left[U + V\frac{\varrho_o}{\varrho_f}\right] \tag{27}$$

$$\frac{C_f}{C_o} \approx \left[1 + \frac{C^2 + C_1 C_2}{\nu} + \frac{C_2}{\mu}\right]^{-1} \tag{28}$$

with $C_2 = 0.375$.

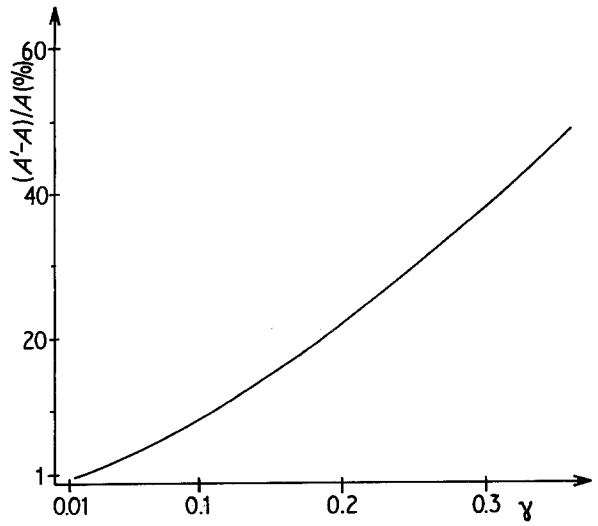

Figure 4 Relative deviation of A' (Equation 22) from exact A (Equation 8) with γ (Equation 5). The value of b is irrelevant ($-10 < b < 100$).

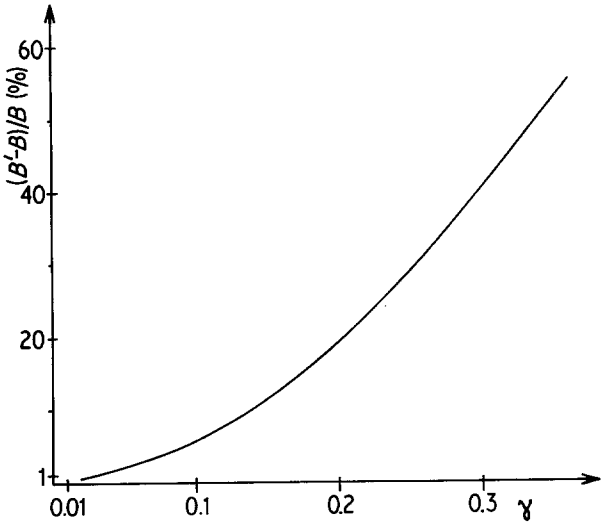

Figure 5 Relative deviation of B' (Equation 23) from exact B (Equation 9) with γ (Equation 5). The value of b is irrelevant ($-10 < b < 100$).

3.3. Comparison with previous expressions obtained for large thickness

In the case of monocrystalline films, taking $D_g = a$, the following approximate expression has been empirically proposed [1, 14]:

$$\frac{\varrho_{fm}}{\varrho_o} \approx 1 + k^{-1}\left[0.36 \ln\frac{1}{p} + 1.144 \ln\frac{1}{t}\right] \tag{29}$$

provided that $k \gg 0.01$, $t > 0.3$, $p > 0.3$, where $k = a/\lambda_0$. Equation 29 can be compared with Equation 24 for

$$C_1 = -C$$

to give

$$\varrho_{fm}/\varrho_o \approx 1 + \nu^{-1} + 0.375\mu^{-1} \tag{30}$$

in qualitative agreement with Equation 29.

In the case of polycrystalline films the following asymptotic formula has been proposed [1, 15]:

$$\frac{\varrho_{fp}}{\varrho_g} \approx 1 + \mu^{-1}[4.7\nu^{-1} + 3]^{-1} \tag{31}$$

whose validity range is $\mu > 0.1$, $0.1 < \nu < 4$, whereas Equation 24 gives

$$\frac{\varrho_{fp}}{\varrho_g} \approx 1 + \frac{C_2}{\mu}\frac{1}{1 + (C^2 + C_1 C_2)/\nu} \tag{32}$$

with $C_1 = 1 - C$, i.e.

$$\frac{\varrho_{fp}}{\varrho_g} \approx 1 + \mu^{-1}\frac{1}{2.62 + 4\nu^{-1}} \tag{33}$$

which is not far from the empirical Relation 31, whose validity range differs.

In the case of an infinitely thick polycrystalline film, Equation 24 takes the form

$$\frac{\varrho_g}{\varrho_o} \approx 1 + \frac{C^2 + C_1 C_2}{\nu} \tag{34}$$

with $C_1 = 1 - C$ to give

$$\varrho_g/\varrho_o \approx 1 + 1.5\nu^{-1} \tag{35}$$

whereas the expression obtained [1, 16] under the

assumption of isotropic grain-boundary scattering is

$$\varrho_g/\varrho_o = 1 + 1.45v^{-1} \quad (36)$$

in good agreement with Equation 35.

When no grain-boundary scattering occurs Equation 24 becomes

$$\varrho_f/\varrho_o \approx 1 + 0.375\mu^{-1} \quad (37)$$

in good agreement with the usual Fuchs–Sondheimer asymptotic expression [1, 7]

$$\left.\frac{\varrho_f}{\varrho_o}\right|_{FS} \approx 1 + \tfrac{3}{8}(1 - p)k^{-1} \quad (38)$$

since the deviation of $\ln(1/p)$ from $(1 - p)$ is not marked as long as p does not take low values.

3.4. Approximate expressions at low thickness or grain size

When Equation 22 and 23 are valid, Equations 1, 2, 3, 10 and 13 becomes

$$\frac{\sigma_f}{\sigma_o} \approx \frac{3}{2b}\ln\gamma^{-1}[1 - (2\ln\gamma^{-1})^{-1}] \quad (39)$$

$$\frac{\beta_f}{\beta_o} \approx (b\gamma\ln\gamma^{-1})^{-1}(1 - 2\gamma) \quad (40)$$

$$\frac{R_{Hf}}{R_{Ho}} \approx \tfrac{2}{3}\gamma^{-1}(\ln\gamma^{-1})^{-2}(1 - 2\gamma) \quad (41)$$

$$S_f \approx -\frac{\pi^2 B^2 T}{3eE_F}[U + V(b\gamma\ln\gamma^{-1})^{-1}(1 - 2\gamma)] \quad (42)$$

$$C_f \approx \frac{3LT\ln\gamma^{-1}}{2}\frac{}{b}[1 - (2\ln\gamma^{-1})^{-1}] \quad (43)$$

When no grain-boundary scattering occurs Equations 39 to 41 take the following forms at very low thickness:

$$\frac{\sigma_f}{\sigma_o} \approx \tfrac{3}{2}\mu\ln\mu^{-1} \qquad \mu \ll 1 \quad (44)$$

$$\frac{\beta_f}{\beta_o} \approx (\ln\mu^{-1})^{-1} \qquad \mu \ll 1 \quad (45)$$

$$\frac{R_{Hf}}{R_{Ho}} \approx \tfrac{2}{3}\mu^{-1}(\ln\mu^{-1})^{-2} \qquad \mu \ll 1 \quad (46)$$

Equations 44 to 46 have previously been obtained [17] in the framework of the extended Cottey conduction model [1, 18].

In the case of very thin monocrystalline films the grain diameter D_g takes values independent of the film thickness; consequently v = constant and $\mu \ll 1$. Hence

$$b \approx \mu^{-1} \quad (47)$$

$$\gamma \approx (1 + C^2v^{-1}) \quad (48)$$

$$\ln\gamma^{-1} \approx \ln\mu^{-1} \quad (49)$$

and Equations 39 to 41 become

$$\frac{\sigma_f}{\sigma_o} \approx \tfrac{3}{2}\mu\ln\mu^{-1} \qquad \mu \ll 1, D_g = \text{constant} \quad (50)$$

$$\frac{\beta_f}{\beta_o} \approx [(1 + C^2v^{-1})\ln\mu^{-1}]^{-1} \qquad \mu \ll 1, D_g = \text{constant} \quad (51)$$

$$\frac{R_{Hf}}{R_{Ho}} \approx \tfrac{2}{3}\mu^{-1}(1 + C^2v^{-1})^{-1}(\ln\mu^{-1})^{-2} \qquad \mu \ll 1, D_g = \text{constant} \quad (52)$$

It appears that the electrical conductivity exhibits the same behaviour whether grain-boundary scattering exists or not. On the other hand, the values of the t.c.r. and the Hall coefficient are lower than those obtained in the absence of grain-boundary scattering.

The case of very thin continuous polycrystalline films is related to the fact that the thickness threshold for the continuity of the film generally corresponds to the grain size [1, 9]; hence this case is in close analogy with that of a monocrystalline film where the grain diameter is equal to the film thickness. The case is the same for very thin columnar films. Hence

$$b \approx k^{-1}[M(p) - CN(t)] \qquad k \ll 1 \quad (53)$$

with

$$M(p) = \begin{cases} \ln 1/p & p > 0.3 \\ \dfrac{2(1 - p)}{1 + p} & 0 \leqslant p \leqslant 1 \end{cases} \quad (53')$$

$$N(t) = \begin{cases} \ln 1/t & t > 0.3 \\ \dfrac{2(1 - t)}{1 + t} & 0 \leqslant t \leqslant 1 \end{cases} \quad (53'')$$

$$\gamma \approx C\{M(p)[CN(t)]^{-1} - 1\}^{-1} \qquad k \ll 1 \quad (54)$$

$$A(b, \gamma_o) \approx A_4 k \qquad k \ll 1 \quad (55)$$

$$B(b, \gamma_o) \approx B_4 k^2 \qquad k \ll 1 \quad (56)$$

with

$$A_4 = A(1, \gamma_o)[M(p) - CN(t)]^{-1} \quad (57)$$

$$B_4 = B(1, \gamma_o)[M(p) - CN(t)]^{-2} \quad (58)$$

Consequently

$$\frac{\sigma_f}{\sigma_o} \approx A_4 k \qquad k \ll 1, D_g = a \quad (59)$$

$$\frac{\beta_f}{\beta_o} \approx \frac{B_4}{A_4}k^2 \qquad k \ll 1, D_g = a \quad (60)$$

$$\frac{R_{Hf}}{R_{Ho}} \approx \frac{B_4}{A_4^2} \qquad k \ll 1, D_g = a \quad (61)$$

Since

$$B(1, \gamma_o) \approx A^2(1, \gamma_o) \qquad \gamma_o > 2 \quad (62)$$

new equations can be derived from Equations 57, 58 and 62:

$$R_{Hf}/R_{Ho} \approx 1 \qquad k \ll 1, \gamma_o > 2, D_g = a \quad (63)$$

$$R_{Hf}/R_{Ho} \approx B(1, \gamma_o)[A(1, \gamma_o)]^{-2} \qquad k \ll 1, \gamma_o < 2 \quad (64)$$

As shown in Table I, marked variations in R_{Hf}/R_{Ho}

may be observed when γ_o varies, i.e. for variations in t and/or p.

For polycrystalline films of given thickness with very fine grains $v \ll 1$ and therefore

$$b \approx (1 - C)v^{-1} \qquad v \ll 1 \tag{65}$$

$$\gamma \approx C^2(1 - C)^{-1} = \gamma_1 \tag{66}$$

where $\gamma_1 = -5.8$. Consequently

$$\sigma_f/\sigma_o \approx A_5 v \qquad v \ll 1 \tag{67}$$

$$\beta_f/\beta_o \approx B_5 v^2 \qquad v \ll 1 \tag{68}$$

$$R_{Hf}/R_{Ho} \approx B_5/A_5^2 \qquad v \ll 1 \tag{69}$$

where

$$A_5 = (1 - C)^{-1}A(1, \gamma_1) \tag{70}$$

$$B_5 = (1 - C)^{-2}B(1, \gamma_1) \tag{71}$$

Numerical values of $A(1, \gamma)$ and $B(1, \gamma)$ (Table I) show that for $\gamma = \gamma_1$

$$B(1, \gamma_1) \approx [A(1, \gamma_1)]^2 \tag{72}$$

Therefore

$$R_{Hf}/R_{Ho} \approx 1 \qquad v \ll 1 \tag{73}$$

4. Some physical consequences

Equation 24 expresses the Matthiessen's rule [1, 11] which assumes the additivity of the contributions to the electrical resistivity due to the various sources of scattering. The earliest empirical formulation in this form was proposed several years ago [19].

Equation 24 can be rewritten in the following form:

$$\frac{\varrho_f}{\varrho_1} \approx 1 + C_2 \frac{\varrho_o}{\varrho_1} \frac{1}{\mu} \tag{74}$$

with

$$\varrho_1 = \varrho_o\left(1 + \frac{C^2 + C_1C_2}{v}\right) \tag{75}$$

Introducing

$$\lambda_1 = \lambda_o \frac{\varrho_o}{\varrho_1} \tag{76}$$

and

$$\mu_1 = \frac{\lambda_o}{\lambda_1}\mu \tag{77}$$

Equation 74 becomes

$$\frac{\varrho_f}{\varrho_1} \approx 1 + \frac{C_2}{\mu_1} \tag{78}$$

This equation has the same form as the usual Fuchs–Sondheimer expression (Equation 37) and can be called the effective F–S equation, as previously suggested [1, 20]; λ_1 is then an effective mean free path [20, 21].

In the case of polycrystalline films, one can refer to the transport parameters of the infinitely thick film (subscript g); one then obtains

$$\frac{\varrho_{fp}}{\varrho_g} \approx 1 + \frac{C_2}{\mu_g} \tag{79}$$

and

$$\lambda_g = \frac{\lambda_o \varrho_o}{\varrho_g} \tag{80}$$

as expected [1] since the effective parameter μ_g can be defined by

$$\mu_g = \frac{\lambda_o}{\lambda_g}\mu \tag{81}$$

Comparing Equation 26 with Equation 24 shows that the size effect in the Hall coefficient is less marked than in the resistivity of a metal film of polycrystalline columnar or monocrystalline structure, at large reduced thickness. This feature has been separately established for any structure [2, 4] and confirmed by many experiments [1].

In the case where the grain diameter remains constant at low film thickness the variation in the conductivity (Equation 50) is not altered by the grain-boundary scattering whereas this is the case for the t.c.r. (Equation 51) and the Hall coefficient (Equation 52).

From Equation 52, it is easily seen that the ratio of the reduced Hall coefficient of monocrystalline films to that of the monocrystal sample $R_{Hf}/R_{Ho}|_{FS}$, may be written as

$$R_{Hf}/R_{Ho} = R_{Hf}/R_{Ho}|_{FS}\,(1 + C^2v^{-1})^{-1} \tag{82}$$

This equation suggests a procedure for an experimental determination of v. It also shows that the variations in R_{Hf}/R_{Ho} have a lower magnitude in the presence of grain-boundary scattering.

From Equation 50 and 52 it is deduced that the size effect in the Hall coefficient is less marked than in the electrical resistivity at low thickness, as expected since theoretical studies [12] have also established this feature at large thickness.

Equation 51 shows that the t.c.r. cannot take negative values at low thickness if the grain size remains constant.

At low film thickness, when the grain diameter is equal to the film thickness, comparing Equations 50 and 59 suggests that the size effect in the electrical conductivity is more marked for $D_g = a$. In this case the Hall coefficient can take values near unity, whereas this is not the case for a constant value of D_g.

One must not forget that a nucleation-growth of thin metal films often occurs with $D_g = a$ [1, 3] except in the cases of chemically deposited films [22–25] which can be built in clusters [26, 27].

As expected when a polycrystalline film contains very fine grains, the grain-boundary scattering is of major importance and determines all the transport properties (Equation 67 to 69); moreover the Hall coefficient is close to that of the bulk material.

Since the Hall coefficient of a thin metal film could take in some cases the value unity at both large and low thickness, we have calculated an asymptotic expression of the Hall coefficient for $\gamma \gg 1$ which is more accurate than Equation 26.

Introducing the parameter G as

$$G = 1 + C^2v^{-1} \tag{83}$$

in Equations 8 and 9 gives

$$A(b, \gamma) = \frac{1}{G}\gamma\left[\gamma - \tfrac{1}{2} + (1 - \gamma^2)\ln(1 + \gamma^{-1})\right] \quad (84)$$

$$B(b, \gamma) = \frac{1}{G^2}\gamma^2\left[\gamma^{-1} - 2 + 2\gamma\ln(1 + \gamma^{-1})\right] \quad (85)$$

For large values of γ the asymptotic expressions for Equations 84 and 85 are

$$A(b, \gamma) \approx \frac{1}{G}\left(1 - \frac{3}{8\gamma} + \frac{1}{5\gamma^2}\right) \quad (86)$$

$$B(b, \gamma) \approx \frac{1}{G^2}\left(1 - \frac{3}{4\gamma} + \frac{3}{5\gamma^2}\right) \quad (87)$$

Hence

$$\frac{R_{Hf}}{R_{Ho}} \approx 1 + \frac{1}{\gamma^2}\left(\tfrac{1}{5} - \tfrac{9}{64}\right) \quad (88)$$

In the absence of grain-boundary scattering γ reduces to μ and Equation 88 is identical to a previous result [17].

In the case of infinitely thick polycrystalline films, γ takes the asymptotic value γ_g given by

$$\gamma_g = \frac{\nu}{1 - C}(1 + C^2\nu^{-1}) \quad (89)$$

It is clear that

$$|\gamma_g| > \frac{C^2}{C - 1} = 5.8 \quad (90)$$

and consequently Equation 88 gives

$$1 < R_{Hg}/R_{Ho} < 1 + 1.7 \times 10^{-3}$$

i.e

$$R_{Hg}/R_{Ho} \approx 1 \quad (91)$$

in good agreement with direct tabulations [12].

5. Discussion

The total set of Equations 1 to 9 is valid as long as the extended Cottey model can be used for calculating the effect of electron scattering at the film surface. Recent discussions [28, 29] have established that this is the case if the electronic reflection coefficient p takes values higher than 0.3; a similar condition is required for the electronic transmission coefficient t [1], but no limitation due to the film thickness holds, as recently pointed out [28].

The approximate equations which are proposed in this paper can be replaced by other approximate equations if the validity range is not convenient for practical implementation; empirical suggestions have recently been made in this way [4].

From a general point of view, it is seen that the electrical resistivity and its temperature coefficient are basic tools for analysing the transport properties; no further information can be derived from Hall measurements, which can only ensure the total consistency of the assumptions. Thermoelectric measurements depend on parameters (U and V) which can be determined by simultaneous experiments on conductivity and t.c.r., as previously illustrated in the case of polycrystalline and monocrystalline films [10].

For separating polycrystalline effects from columnar effects, the only way consists in calculating from experiments the coefficient C_1, whose value is -0.27 for polycrystalline film and -1.27 for columnar films; however, since C_1 is multiplied by the unknown quantity $N(t)$ in the linearized expressions, this path does not markedly differ from a blind alley. The equations could be used for the calculation of t, provided that the structure was well known, for instance from crystallographic studies.

6. Conclusion

Approximate expressions may be used for expressing the whole set of transport properties of thin metal films, in the framework of bidimensional and three-dimensional conduction models. The simplest linearized expressions are based on the expression for the electrical conductivity only. Further simple linearized expressions can be proposed in the rest of the experimental domain.

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Received 13 May
and accepted 12 June 1985