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Effects of electron scatterings on thermal conductivity of thin metal films

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In the framework of the statistical conduction models for polycrystalline and monocrystalline metal films, the thermal conductivity due to electron transport is calculated under the further assumption that the electron relaxation time in the bulk material depends on electron energy. It is shown that the same function describes the size effect in thermal and electrical conductivities, and that the Wiedemann–Franz law holds for any electron scattering in thin metal films.

1. Introduction

Although it is well known [1] that the transport of heat plays an important role in determining the kinetics of nucleation and growth of thin films, very few workers [1–5] have proposed theoretical expressions for the thermal conductivity of thin films and none of these took into account the effect of electronic scattering at grain boundaries (it is suggested in [5]). Moreover, an extended knowledge of the variations in thermal conductivity of superimposed layers would be convenient for solving problems involving many efficient multilayer insulations used, for instance, for the storage of cryogenic propellants.

In view of these basic and technical interests, we propose in this paper an analysis of the thermal conductivity of thin metal films, starting from the models [6, 7] which have recently been presented for representing the effects of simultaneous background, external-surface and grain-boundary scatterings of conduction electrons.

2. Theoretical study

The general expression for the current density, J_x, and heat flux, $\mathscr{U}_x$, in a thin metal film subjected to an electric field, E_x, and a temperature gradient, $\partial T/\partial x$, in the x-direction are [2]

$$J_x = -2e\left(\frac{m}{h}\right)^3 \int \mathscr{F}_1 v_x d^3 v \tag{1}$$

and

$$\mathscr{U}_x = 2\left(\frac{m}{h}\right)^3 \int \mathscr{F}_1(\epsilon - \epsilon_F) v_x d^3 v, \tag{2}$$

respectively, where e is the absolute electron charge, m is the electron mass, v_x is the x-component of electron velocity v, ϵ is the electron energy, ϵ_F is the Fermi energy, $\mathscr{F}_1$ is the deviation from the equilibrium distribution function, $\mathscr{F}_0$, and h is Planck's constant.

$\mathscr{F}_1$ obeys the Boltzmann equation for charge transport, i.e., in polar co-ordinates (r, θ, ψ), [2]

$$\mathscr{F}_1 = \frac{e}{m}\tau(\theta)\frac{\partial \mathscr{F}_0}{\partial v_x}\left[E'_x + \frac{1}{e}\frac{\epsilon - \epsilon_F}{T}\frac{\partial T}{\partial x}\right], \tag{3}$$

where $\tau(\theta)$ is the electron relaxation time in the film and E'_x is the effective electric field, given by [2]

$$E'_x = E_x + \frac{1}{e}\frac{\partial \epsilon_F}{\partial x}. \tag{4}$$

In the tridimensional [8] and bidimensional [7] conduction models which describe electronic conduction in polycrystalline and monocrystalline films, respectively, the electron mean free paths,

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$\lambda_p(\theta)$ and $\lambda_m(\theta)$, respectively, are given by [8, 7]

$$[\lambda_p(\theta)]^{-1} = \lambda_0^{-1} + C^2 D_g^{-1} \ln \frac{1}{t} + \left[d^{-1} \ln \frac{1}{p} + (1-C) D_g^{-1} \ln \frac{1}{t} \right] |\cos\theta| \tag{5}$$

and

$$[\lambda_m(\theta)]^{-1} = \lambda_0^{-1} + C^2 D_g^{-1} \ln \frac{1}{t} + \left[d^{-1} \ln \frac{1}{p} - C D_g^{-1} \ln \frac{1}{t} \right] |\cos\theta|, \tag{6}$$

where λ_0 is the bulk mean free path, D_g is the average grain diameter (D_g is a constant for polycrystalline films; $D_g \geqslant d$ for monocrystalline films), d is the film thickness, p is the electronic specular reflection coefficient at the external film surface [1], t is the electronic transmission coefficient at the grain boundary [9] and C is a constant ($C = 4/\pi$ [6, 7]).

The bulk mean free path, λ_0, can be expressed from the bulk relaxation time, τ_0 [2] as

$$\lambda_0 = v_F \cdot \tau_0(\epsilon_F) \tag{7}$$

and the energy dependence of λ_0 then results from the energy dependences of v and λ_0.

It is assumed that the free electron model is valid, i.e. that

$$v \sim \epsilon^{1/2}. \tag{8}$$

The energy dependence of τ_0 is defined by [4]

$$\tau_0 = \tau_b \epsilon^q, \tag{9}$$

where τ_b does not depend on ϵ.

This equation can describe an electronic scattering due to impurities or frozen-in defects [2].

When solving integral equations for J_x and $\mathscr{U}_x$, it is assumed that the electron distribution is slightly perturbed, so that the following expansion (Equation 10 in [2]) is valid

$$-\int g(\epsilon) \frac{\partial \mathscr{F}_0}{\partial \epsilon} d\epsilon = g(\epsilon_F) + \frac{1}{6} (\Pi B T)^2 \left. \frac{\partial^2 g(\epsilon)}{\partial \epsilon^2} \right|_{\epsilon=\epsilon_F}, \quad \text{for } \Pi B T \epsilon_F^{-1} \ll 1, \tag{10}$$

where B is the Boltzmann constant and T is the absolute temperature.

Under the above assumptions, integrating Equations 1 and 2 gives

$$J_x = \frac{4\pi e^2}{m^2} \left(\frac{m}{h}\right)^3 \left[K_0' E_x + \frac{1}{eT} K_1' \frac{\partial T}{\partial x} \right] \tag{11}$$

and

$$\mathscr{U}_x = -\frac{4\pi \epsilon}{m^2} \left(\frac{m}{h}\right)^3 \left[K_1' E_x + \frac{1}{eT} K_2' \frac{\partial T}{\partial x} \right], \tag{12}$$

with

$$K_0' = 2 \left(\frac{2}{m}\right)^{1/2} \tau_b \epsilon_F^{q+3/2} b^{-1} L_0(a), \tag{13}$$

$$K_1' = \frac{(\Pi B T)^2}{3} 2 \left(\frac{2}{m}\right)^{1/2} \tau_b \epsilon_F^{q+1/2} \times \left[\left(q + \frac{1}{2}\right) b^{-2} L_1(a) + b^{-1} L_0(a) \right] \tag{14}$$

and

$$K_2' = \frac{(\Pi B T)^2}{3} K_0', \quad \text{for } \Pi B T \epsilon_F^{-1} \ll 1, \tag{15}$$

and with

$$L_0(a) = a - \frac{1}{2} + (1 - a^2) \ln (1 + a^{-1}) \tag{16}$$

and

$$L_1(a) = -2 + a^{-1} + 2a \ln (2 + a^{-1}). \tag{17}$$

The variables a and b are defined as follows for polycrystalline and monocrystalline films (index p and m, respectively) [8, 7]:

$$a_p = b_p^{-1} \left(1 + C^2 \lambda_0 D_g^{-1} \ln \frac{1}{t} \right), D_g = \text{constant} \tag{18}$$

and

$$b_p = \lambda_0 \left[d^{-1} \ln \frac{1}{p} + (1-C) D_g^{-1} \ln \frac{1}{t} \right] \tag{19}$$

and

$$a_m = b_m^{-1} \left[1 + C^2 \lambda_0 D_g^{-1} \ln \frac{1}{t} \right], \quad D_g \geqslant d \tag{20}$$

and

$$b_m = \lambda_0 \left[d^{-1} \ln \frac{1}{p} - C D_g^{-1} \ln \frac{1}{t} \right]. \tag{21}$$

Since

$$\frac{\partial \mathscr{U}_x}{\partial E_x} = -\frac{1}{T} \frac{\partial J_x}{\partial \left\{ \frac{\partial T}{\partial x} \right\}}, \tag{22}$$

these equations satisfy an essential physical requirement: the Kelvin–Onsager principle.

The thermal conductivty, $\mathscr{C}_{\mathrm{fe}}$, due to free electron transport, is calculated from Equations 11 and 12 by introducing the general definition [2]

$$\mathscr{U} = -\ \mathscr{C}_{\mathrm{fe}}\ \mathrm{grad}\ T. \qquad (23)$$

Further, setting $J_x = 0$ in Equation 11 then gives

$$\mathscr{C}_{\mathrm{fe}} = \frac{\Pi^2 B^2 T}{3e^2}\left[\frac{4\Pi e^2}{m^2}\left(\frac{m}{\hbar}\right)^3 2\lambda_0 \epsilon_{\mathrm{F}} b^{-1} L_0(a)\right],$$

$$\text{for } \Pi B T \epsilon_{\mathrm{F}}^{-1} \ll 1. \qquad (24)$$

Since the film conductivity σ_{f}, as shown in previous papers [6, 7], is given by

$$\sigma_{\mathrm{f}} = \frac{4\Pi e^2}{m^2}\left(\frac{m}{h}\right)^3 2\lambda_0 \epsilon_{\mathrm{F}} b^{-1} L_0(a) \qquad (25)$$

then

$$\mathscr{C}_{\mathrm{fe}} = \frac{\pi^2 B^2 T}{3e^2}\sigma_{\mathrm{f}}. \qquad (26)$$

3. Physical point of view

We then conclude that the size effects in thermal conductivity and electrical conductivity are identical, whatever the value of the average grain diameter and whether or not it is constant (polycrystalline films) or equal to the film thickness, or larger (monocrystalline films).

Since the multidimension conduction models [6, 7] reduce to the Fuchs–Sondheimer conduction model [1] when the effects of grain boundaries are negligible [6, 7], Equation 26 is also valid in the Fuchs–Sondheimer model. This conclusion is in agreement with the results of Verma and Jain who studied correlated size effects in electrical and thermal conductivities in the Fuchs–Sondheimer model, assuming that τ_0 is not energy dependent [3]; they further proposed an approximate expression for thermal conductivity [4] when τ_0 depends on energy.

The value of the Wiedemann–Franz ratio, $\mathscr{R}_{\mathrm{WF}}$, [2]

$$\mathscr{R}_{\mathrm{WF}} = \frac{\mathscr{C}_{\mathrm{fe}}}{\sigma_{\mathrm{f}} T} \qquad (27)$$

is then calculated from Equation 26

$$\mathscr{R}_{\mathrm{WF}} = \Pi^2 B^2 / 3e^2. \qquad (28)$$

It is seen that $\mathscr{R}_{\mathrm{WF}}$ is equal to the standard Lorenz number [2].

We then conclude that the Lorenz number expresses the Wiedemann–Franz ratio when the conduction electrons are scattered by phonons, grain broundaries, external surfaces and impurities or defects.

These results agree with the general theoretical predictions of Ziman [2]; for the validity of this calculation it must be assumed that electron scattering is elastic, so that the Boltzmann equation reduces to an integral equation over a single constant-energy surface.

In our opinion, this condition is not clearly satisfied in the case of the multidimensional conduction models [6–8] since they are based on the definition of a mean free path in the film which depends on the fraction of electrons which are elastically scattered, which, in turn, is dependent on p and t. However, it can be suggested that the p- and t-dependent mean free path is a mathematical tool which implicitly expresses both the boundary conditions at film surfaces and grain boundaries and the effects of background and impurities scatterings. Cottey [10] has shown such an equivalence for external-surface scattering but the general case has never been treated.

4. Conclusion

It is seen from theoretical studies of the thermal electronic conductivity in the framework of statistical multidimensional conduction models [6, 7] that the Wiedemann–Franz law holds in any case of electronic scattering in thin metal films at room temperature.

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