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SIZE EFFECTS IN SOME TRANSPORT COEFFICIENTS FOR DOUBLE-LAYER THIN METALLIC FILMS

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A theoretical analysis, based on the Fuchs-Sondheimer formalism, is given for the transport coefficients — electrical conductivity, thermal conductivity, thermoelectric power and Peltier coefficient — of double-layer thin metallic films. Two different relaxation times are assumed for the bulk scattering of the conduction electrons in the layers. The relaxation times are taken as functions of energy, $\tau_1 \sim \varepsilon^\alpha$ and $\tau_2 \sim \varepsilon^\beta$ for the lower (base) and the upper layers, respectively. The electron scattering on surfaces or interface is described by the parameters P, Q. The parameters P (P_{10}, P_{20}, P_{12}, P_{21}) are the Fuchs parameters of the specular reflection on the three surfaces (two outer surfaces and the interface between the layers). The parameter Q corresponds to the fraction of the conduction electrons refracted at the interface. A numerical calculation was made for the electrical conductivity and thermoelectric power in the case of a total transmission of the conduction electrons at the interface between the layers.

РАЗМЕРНЫЕ ЭФФЕКТЫ В ТРАНСПОРТНЫХ КОЭФФИЦИЕНТАХ ДЛЯ ДВУХСЛОЙНЫХ ТОНКИХ МЕТАЛЛИЧЕСКИХ ПЛЕНОК

В работе на основе формализма Фухса–Зондгеймера проведен теоретический анализ транспортных коэффициентов – электропроводности, удельной теплопроводности, термоэлектрического напряжения и коэффициента Пельтье – для двухслойных металлических пленок. Анализ сделан в предположении, что для объемного рассеяния электронов проводимости в слоях существуют два различных времени релаксации. Предполагается, что время релаксации зависит от энергии по формулам $\tau_1 \sim \varepsilon^\alpha$ и $\tau_2 \sim \varepsilon^\beta$ для нижней (базисной) и для верхней пленок соответственно. Рассеяние электронов на поверхности пленки и на разделе слоев описано при помощи параметров P, Q, которые представляют собой параметры Фухса для зеркальных отражений на трех поверхностях (две внешние поверхности и раздел обоих слоев) и долю электронов проводимости, которые изменили направление на границе раздела обоих слоев. Проведен также анализ численных расчетов электропроводимости и термоэлектрического напряжения в случае, когда электроны проводимости полностью проходят через границу раздела обоих слоев.

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I. INTRODUCTION

When the thickness of a metal film is comparable to the bulk mean free path of the conduction electrons, the surface of the film contributes significantly to the scattering of electrons. This leads to the size effects. The analysis of the size effects was carried out by Fuchs [1], Sondheimer [2] and many others by solving the Boltzmann transport equation with appropriate boundary conditions. An extension of their model was made, e.g., by Lucas [3, 4] who introduced two specularity parameters P_1 and P_2 to characterize surface scattering at the outer surfaces of double-layer thin metallic films. Lucas assumed that the internal interface between the layers does not cause any reflection or additional scattering of the conduction electrons. The average longitudinal electrical conductivity of double-layer thin metallic films was calculated in [5], where the scattering at the interface between the layers was taken into account. The authors in [5] used simple boundary conditions which were a generalization of the well-known Fuchs boundary conditions. They confined their calculation to the simplest case when the film was subjected only to an external electric field applied in the direction parallel to the surface. Verma and Jain [6, 7] studied the size effects relating to transport coefficients in a single metallic film within the framework of the Fuchs theory. Their analysis concerns a single film which is subjected to an electric field and a temperature gradient parallel to the film surface. They deal with a special case where both surfaces had the same specularity parameter. Later the transport coefficients for this model were recalculated [8, 9] for the case when the two surfaces have different specularity parameters.

The purpose of this paper is to study the transport coefficients of double-layer thin metallic films taking into account the films subjected to an external electric field $\mathscr{E}_x$ and a temperature gradient ∇T in the x-direction parallel to the film surface. The theoretical analysis in this paper utilizes two relaxation times with the energy dependence $\tau_1 \sim \varepsilon^{\alpha}$ and $\tau_2 \sim \varepsilon^{\beta}$ for the lower (base) and the upper layers, respectively. The values of α and β depend on the predominant scattering mechanism (α, $\beta = -0.5$ for lattice scattering, α, $\beta = 1.5$ for ionized impurity scattering and α, $\beta = 0$ for neutral defects scattering). For simplicity we shall investigate only the case when $\alpha = \beta$.

II. EXPRESSIONS FOR TRANSPORT COEFFICIENTS IN A BULK METAL

For a bulk metal subjected to an external electric field $\mathscr{E}_x$ and a temperature gradient ∇T in the x-direction, the current density J_x and the heat flux U_x are given by the expressions:

$$J_x = -2e\left(\frac{m}{h}\right)^3 \iiint f v_x \, d^3 v \,, \tag{1}$$

$$U_x = 2\left(\frac{m}{h}\right)^3 \iiint f(\varepsilon - \zeta)\, v_x \, d^3 v \,, \tag{2}$$

where e is the elementary change ($e>0$), m the electronic, mass, v_x the velocity of the conduction electrons in the x-direction, h the Planck constant, f the distribution function of the conduction electrons obtained from the Boltzmann transport equation and f^0 the equilibrium Fermi-Dirac distribution function of the electrons,

$$f^0 = \frac{1}{\exp\{(\varepsilon - \zeta)/k_B T\} + 1} \,, \tag{3}$$

ε is the electronic energy, ζ is the chemical potential, k_B is the Boltzmann constant and T is the absolute temperature. The parameter ζ is a function of the temperature. The current density J_x and the heat flux U_x can be rewritten in their general form, such that

$$J_x = e^2 R_0 \mathscr{E}_x + \frac{e}{T} R_1 \left(-\frac{\partial T}{\partial x}\right), \tag{4}$$

$$U_x = e R_1 \mathscr{E}_x + \frac{1}{T} R_2 \left(-\frac{\partial T}{\partial x}\right). \tag{5}$$

The coefficients R_0, R_1 and R_2 can be obtained from the equations for J_x and U_x. Thus using the basic definitions of the transport coefficients we get the results for the bulk metal as follows:

(i) The bulk electrical conductivity

$$\sigma_B = e^2 R_0 \tag{6}$$

(ii) The bulk thermal conductivity

$$K_{TB} = \frac{R_2}{T} - \frac{R_1^2}{R_2 T} \tag{7}$$

(iii) The bulk thermoelectric power

$$S_B = \frac{1}{eT}\left(\frac{R_1}{R_0}\right) \tag{8}$$

(iv) The bulk Peltier coefficient

$$\Pi_B = \frac{1}{e}\left(\frac{R_1}{R_0}\right). \tag{9}$$

III. EXPRESSIONS FOR TRANSPORT COEFFICIENTS IN DOUBLE-LAYER THIN METALLIC FILMS

Let us consider a double-layer thin metallic film whose surfaces are parallel to the plane $z=0$, subjected to an external electric field $\mathscr{E}_x$ and temperature gradient

∇T in the x-dierction (see Figure 1). The electron distribution function $f = f^0 + g\,(v, z)$ obeys the Boltzmann equation, where $g\,(v, z)$ is the small deviation from equilibrium caused by the external thermodynamic forces, i.e. by $\mathscr{E}_x$ and ∇T.

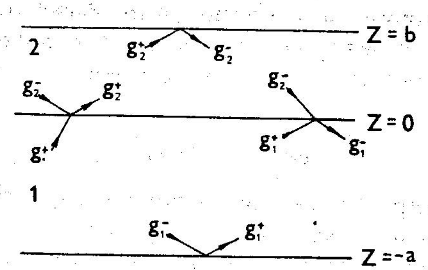


Fig. 1. Reflection and refraction of conduction electrons by the outer layer surfaces and the interface.

According to Bezák and Krempaský [10], the distribution functions g_1^+, g_2^+, g_1^- and g_2^- satisfy the boundary conditions

$$
\begin{aligned}
g_1^+ &= P_{10} g_1^- && \text{at } z = -a \\
g_2^- &= P_{20} g_2^+ && \text{at } z = b \\
g^- &= P_{12} g^+ + Q_{21} g^-{}_2 && \text{at } z = 0 \\
g_2^+ &= P_{21} g_2^- + Q_{12} g_1^+ && \text{at } z = 0,
\end{aligned}
\qquad (10)
$$

where $Q_{12} = Q_{21} = Q$, and the distribution functions g_1^+, g_2^+ and g_1^-, g_2^- are the functions of the conduction electrons with z-components of the positive and negative velocities, respectively. Here 1, 2 refers, respectively, to the lower (base) layer whose thickness is a, and the upper layer whose thickness is b.

The parameters P_{10}, P_{20}, P_{12} and P_{21} are called the Fuchs parameters, they characterize the specularity of the reflection of the conduction electrons from the surfaces and the interface. Because of the roughness of the interface, P_{12} need not be equal to P_{21}. The parameter Q characterizes the probability that an electron is refracted on the interface according to the law of refraction.

The Boltzmann equation for the conduction electrons has the form:

$$v_z \frac{\partial g}{\partial z} + \frac{g}{\tau} = e v_x \frac{\partial f^0}{\partial \varepsilon}\left(E_x + \frac{1}{e}\,\frac{\varepsilon - \zeta}{T}\,\frac{\partial T}{\partial x}\right), \qquad (11)$$

where $E_x = \mathscr{E}_x + \frac{1}{e}\frac{\partial \zeta}{\partial x}$ is the internal electric field.

Solving the Boltzmann transport equations (four equations, i.e. two for each layer) for the conduction electrons with respect to the boundary conditions (10),

we can obtain the average current density $\bar{J}_x$ and average heat flux $\bar{U}_x$ for the double-layer thin metallic film, respectively, by the formulae:

$$
\begin{aligned}
\bar{J}_x = {} & \frac{1}{a+b}\left[a\sigma_{B1} E_x F_1(K, P, Q) + b\sigma_{B2} E_x F_2(K, P, Q)\right] + \\
& + \frac{1}{a+b}\left[\frac{2e}{m_1}\left(\frac{m_1}{h}\right)^3 \left(\frac{2\zeta_1}{m_1}\right)^{1/2} \left(\frac{4\pi a \tau_1}{3m_1}\right)(\pi^2 k_B^2 T^2)\left(\frac{1}{T}\frac{\partial T}{\aleph x}\right)\Psi_1(K, P, Q) + \right. \\
& \left. + \frac{2e}{m_2}\left(\frac{m_2}{h}\right)^3 \left(\frac{2\zeta_2}{m_2}\right)^{1/2} \left(\frac{4\pi b \tau_2}{3m_2}\right)(\pi^2 k_B^2 T^2)\left(\frac{1}{T}\frac{\partial T}{\partial x}\right)\Psi_2(K, P, Q\right].
\end{aligned}
\qquad (12)
$$

$$
\begin{aligned}
\bar{U}_x = {} & \frac{1}{a+b}\left[\frac{2e}{m_1}\left(\frac{m_1}{h}\right)^3 \left(\frac{2\zeta_1}{m_1}\right)^{1/2} \left(\frac{4\pi a \tau_1}{3m_1}\right)(-\pi^2 k_B^2 T^2)\, E_x \Psi_1(K, P, Q) + \right. \\
& \left. + \frac{2e}{m_2}\left(\frac{m_2}{h}\right)^3 \left(\frac{2\zeta_2}{m_2}\right)^{1/2} \left(\frac{4\pi b \tau_2}{3m_2}\right)(-\pi^2 k_B^2 T^2)\, E_x \Psi_2(K, P, Q)\right] + \\
& + \frac{1}{a+b}\left[\left(\frac{a\sigma_{B1}}{3e^2}\right)(-\pi^2 k_B^2 T^2)\left(\frac{1}{T}\frac{\partial T}{\partial x}\right) F_1\, K, P, Q) + \right. \\
& \left. + \left(\frac{b\sigma_{B2}}{3e^2}\right)(-\pi^2 k_B^2 T^2)\left(\frac{1}{T}\frac{\partial T}{\partial x}\right) F_2\,(K, P, Q)\right].
\end{aligned}
\qquad (13)
$$

We have taken into consideration that the layers may be of different metals, so we consider different effective masses m_1, m_2, Fermi velocities v_{F1}, v_{F2}, relaxation lengths L_1, L_2 (or bulk relaxation times $\tau_1 \sim \varepsilon^\alpha$ and $\tau_2 \sim \varepsilon^\beta$) and electronic densities n_1, n_2. The Fermi surface for each layer is assumed to be spherical.

The functions $F_1(K, P, Q)$ and $\Psi_1(K, P, Q)$ are defined as follows:

$$
\begin{aligned}
F_1(K, P, Q) = {} & \left[1 - \frac{3}{4K_1}\int_0^1 \mathrm{d}x_1 (x_1 - x_1^3)\,(1 - A)\,\{(1 - P_{10}) + D^{-1}(1 + P_{10}A) \times \right. \\
& \left. \times (X_1 + CQY_1)\right].
\end{aligned}
\qquad (14)
$$

$$
\begin{aligned}
\Psi_1(K, P, Q) = {} & \left[\left(1 + \frac{2\alpha}{3}\right) - \frac{(\alpha+1)}{K_1}\int_0^1 \mathrm{d}x_1\,(x_1 - x_1^3)\,(1 - A)\left\{(1 - P_{10}) + \right.\right. \\
& \left. + D^{-1}\,(1 + P_{10}A)\left(X_1 + CQY_1\left(1 + \frac{\beta\tau_1/\zeta_2 - \alpha}{2(\alpha - 1)}\right)\right)\right\} - \\
& - \frac{\zeta_1}{2K_1}\int_0^1 \mathrm{d}x_1\,(x_1 - x_1^3)\,\frac{\partial}{\partial \varepsilon}\left((1 - A)\,\{(1 - P_{10}) + \right. \\
& \left.\left. + D^{-1}\,(1 + P_{10}A)\,(X_1 + CQY_1)\}\right)_{\varepsilon = \zeta_1}\right],
\end{aligned}
\qquad (15)
$$

where K_1 is the reduced thickness of the lower (base) layer, $K_1 = a/L_1$. Similarly we have defined the functions $F_2(K, P, Q)$ and $\Psi_2(K, P, Q)$ by changing in the integrals (14), (15) the index 1 (2) by the index 2 (1), the parameter α (β) by β (α), the symbol A (B) by the symbol B (A) and C by C^{-1}.

It is interesting to see how the functions F_1, F_2, Ψ_1 and Ψ_2 behave in the limit of large values of K_1 and K_2 (asymptotic approximation):

$$F_1(K, P, Q) = 1 - \frac{3[1-(P_{10}+P_{12})/2]}{8K_1}, \tag{16}$$

$$F_2(K, P, Q) = 1 - \frac{3[1-(P_{20}+P_{21})/2]}{8K_2}, \tag{17}$$

$$\Psi_1(K, P, Q) = 1 - \frac{[1-(P_{10}+P_{12})/2]}{2K_1}, \tag{18}$$

$$\Psi_2(K, P, Q) = 1 - \frac{[1-(P_{20}+P_{21})/2]}{2K_2}, \tag{19}$$

Here we have used the notations:

$$X_1 = \{1 - P_{12} + AP_{12}(1-P_{10}) - B^2P_{20}(P_{21}(1-P_{12}) + Q^2) + + AB^2P_{20}(Q^2 - P_{12}P_{21})(1-P_{10})\}. \tag{20}$$

$$Y_1 = -(1-B)(1+BP_{20}). \tag{21}$$

$$D = 1 - A^2P_{10}P_{12} - B^2P_{20}P_{21} + A^2B^2P_{10}P_{20}(P_{12}P_{21} - Q^2). \tag{22}$$

The quantities A, B and C are

$$A = \exp\left(-\frac{a}{\tau_1|v_{1z}|}\right) = \exp\left(-\frac{a}{L_1\cos\Theta_1}\right) \tag{23}$$

$$B = \exp\left(-\frac{b}{\tau_2|v_{2z}|}\right) = \exp\left(-\frac{b}{L_2\cos\Theta_2}\right) \tag{24}$$

$$C = \frac{L_2 m_1 v_1}{L_1 m_2 v_2}. \tag{25}$$

We have introduced the angle Θ_i such that $v_{iz} = v x_i$, $x_i = \cos\Theta_i$, $i = 1, 2$. Comparing equations (4), (12) and (5), (13) we find that

$$R_{0f} = \frac{a}{a+b}\frac{\sigma_{B1}}{e^2}F_1(K, P, Q) + \frac{b}{a+b}\frac{\sigma_{B2}}{e^2}F_2(K, P, Q). \tag{26}$$

$$R_{1f} = -\frac{a}{a+b}\left(\frac{m_1}{h}\right)^3\left(\frac{2\zeta_1}{m_1}\right)^{1/2}\left(\frac{8\pi\tau_1}{3m_1^2}\right)(\pi^2k_B^2T^e)\,\Psi_1(K, P, Q) - -\frac{b}{a+b}\left(\frac{m_2}{h}\right)^3\left(\frac{2\zeta_2}{m_2}\right)^{1/2}\left(\frac{8\pi\tau_2}{3m_2^2}\right)(\pi^2k_B^2T^e)\,\Psi_2(K, P, Q). \tag{27}$$

$$R_{2f} = \frac{a}{a+b}\frac{\pi^2k_B^2T^e}{3e^2}\sigma_{B1}F_1(K, P, Q) + \frac{b}{a+b}\frac{\pi^2k_B^2T^e}{3e^2}\sigma_{B2}F_2(K, P, Q). \tag{28}$$

From the relations for R_{0f}, R_{1f} and R_{2f} (f, B represent film and bulk respectively) we obtain the following transport coefficients for the case of the double-layer thin metallic film:

(1) Electrical conductivity

$$\sigma_f = \frac{a}{a+b}\sigma_{B1}F_1(K, P, Q) + \frac{b}{a+b}\sigma_{B2}F_2(K, P, Q). \tag{29}$$

(2) Thermal conductivity

$$K_f = \frac{a}{a+b}K_{TB1}F_1(K, P, Q) + \frac{b}{a+b}K_{TB2}F_2(K, P, Q). \tag{30}$$

(3) Thermoelectric power

$$S_f = \frac{S_{B1}\Psi_1(K, P, Q)\Big/\left(\frac{2\alpha}{3}+1\right) + \frac{b}{a}CS_{B2}\Psi_2(K, P, Q)\Big/\left(\frac{2\beta}{3}+1\right)}{F_1(K, P, Q) + \frac{b}{a}CF_2(K, P, Q)} \tag{31}$$

(4) Peltier coefficient

$$\Pi_f = \frac{\Pi_{B1}\Psi_1(K, P, Q)\Big/\left(\frac{2\alpha}{3}+1\right) + \frac{b}{a}C\Pi_{B2}\Psi_2(K, P, Q)\Big/\left(\frac{2\beta}{3}+1\right)}{F_1(K, P, Q) + \frac{b}{a}CF_2(K, P, Q)}. \tag{32}$$

IV. NUMERICAL RESULTS

We will consider a model (special case of the double-layer thin metallic film), wh[illegible] there is no scattering of the [illegible] at the internal interface

For this model the functions $F_1(K, P, Q)$, $F_2(K, P, Q)$, $\Psi_1(K, P, Q)$ and $\Psi_2(K, P, Q)$ are defined as follows:

$$F_1(K, P, Q)=\left[1-\frac{3}{4K_1}\int_0^1 dx(x-x^3)(1-A^2)(1-C(1-B))\right] \quad (33)$$

$$F_2(K, P, Q)=\left[1-\frac{3}{4K_2}\int_0^1 dx(x-x^3)(1-B)(2-A^2(1-B)- -C^{-1}(1-A^2))\right]. \quad (34)$$

$$\Psi_1(K, P, Q)=\left[\left(1+\frac{2\alpha}{3}\right)-\frac{(\alpha+1)}{K_1}\int_0^1 dx(x-x^3)(1-A^2)\times \times(1-C(1-B)\left(1+\frac{\beta-\alpha}{2(\alpha+1)}\right)\right)+\frac{(2\alpha+1)}{2}\times \times\int_0^1 dx(1-x^2)A^2(1-C(1-B))- -\frac{(2\beta+1)b}{4a}\int_0^1 dx(1-x^2)B(1-A^2)\right]. \quad (35)$$

$$\Psi_2(K, P, Q)=\left[\left(1+\frac{2\beta}{3}\right)-\frac{(\beta+1)}{K_2}\int_0^1 dx(x-x^3)(1-B)(2- -A^2(1-B)-C^{-1}(1-A^2)\left(1+\frac{\alpha+\beta}{2(\beta+1)}\right)\right)+ +\frac{(2\beta+1)}{4}\int_0^1 dx(1-x^2)B(2-2A^2(1-B)-C^{-1}(1-A^2))- -\frac{(2\alpha+1)}{2b}\int_0^1 dx(1-x^2)A^2(1-B)(1-C(1-B))\right]. \quad (36)$$

The dependences of the conductivity ratio σ_f/σ_{B1} and the thermoelectric power ratio S_f/S_{B1} on the thickness ratio b/a for $L_1/L_2=10$ and $K_1=10, 5, 1, 0.5$ were calculated using a computer. The results are shown in Figures (2, 3, 4). These dependences were calculated for the three scattering mechanisms: scattering of the conduction electrons by latt[illegible]nons, ionized impurities and neutral defects i.e.

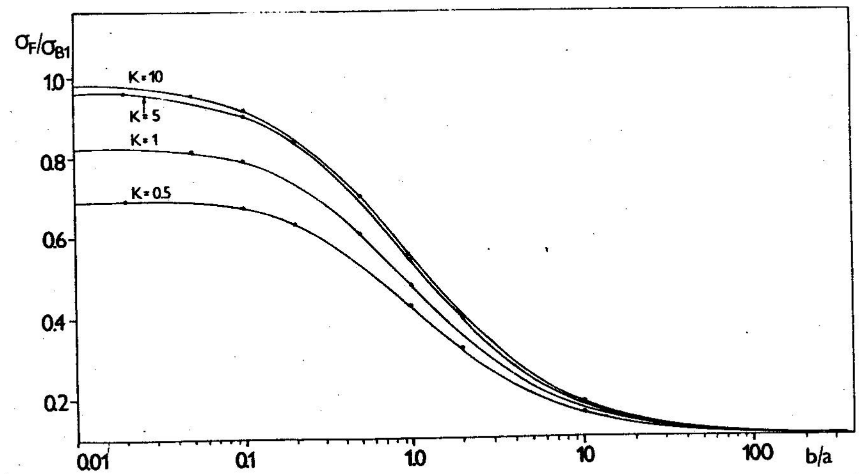
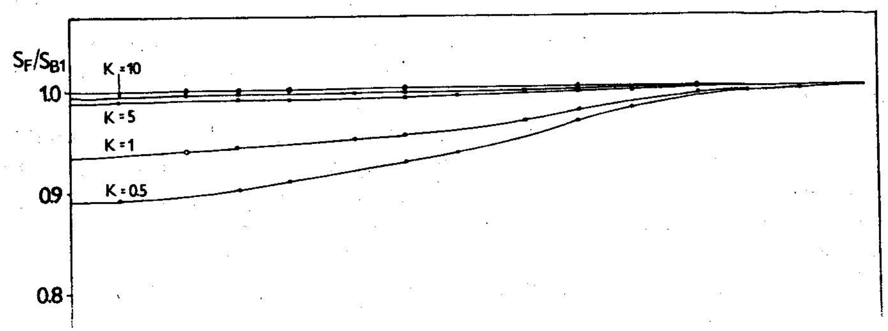


Fig. 2. Variation of σ_F/σ_{B1} with b/a for $K_1=10, 5, 1, 0.5$ and $L_1/L_2=10$.

(b) For very large values of the upper layer thickness ($b\rightarrow\infty$) σ_{B2}/σ_{B1} and S_{B2}/S_{B1} tends to L_2/L_1 and unity, respectively, for $K_1\gg 1$ as well as for $K_1\ll 1$.

The model can be viewed for instance as a rough physical realization of an amalgamated gold film, where the lower (base) and upper layers represent the pure gold layer and the amalgamated layer, respectively. Therefore the gold layer is a reliable detector of mercury. This follows from the remarkable influence of the amalgamated (upper-layer) on the transport coefficients of the film.



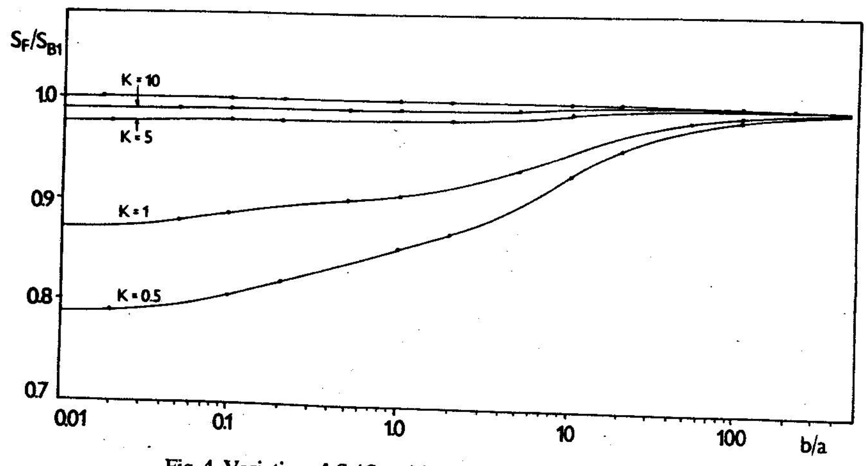


Fig. 4. Variation of S_F/S_{B1} with b/a for the values $\alpha=\beta=1.5$.

V. CONCLUSION

By the above analysis we have obtained general as well as asymptotic expressions for the transport coefficients of a double-layer thin metallic film, taking into account that the film is subjected to an external field $\mathscr{E}_x$ and a parallel temperature gradient ∇T. The analysis has been carried out using a general energy dependence of the relaxation time in the two layers of the film. We have generalized the results obtained by Bezák et al. [5] and have calculated also the thermal conductivity, thermoelectric power and Peltier coefficient for a double-layer thin metallic film. The results show that for a double-layer thin metallic, film, the transport coefficients exhibit size effects, except for the thermoelectric power and the Peltier coefficient if the predominant scattering of the conduction electrons is by lattice phonons. This has a simple physical explanation, namely, if $\alpha=\beta=-0.5$, we have a constant bulk mean free path independent of energy. The analysis has shown that the ratio of the film to bulk electronic thermal conductivity in metals behaves in the same manner as the analogical ratio for the electrical conductivity. They are independent of the type of scattering which is dominating. The same is valid for the Peltier coefficient and thermoelectric power, but they have a strong dependency on the type of the dominating scattering. From variations of the film electrical [illegible]

however, the theory will become somewhat more complicated because of the usual presence of some surface change at the interfaces, the bending of the bands and the possibility of strong charge fluctuations.

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