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THERMAL CONDUCTIVITY OF METALLIC WIRES

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The effect of radial thickness on the thermal conductivity of a free standing wire is investigated. The thermal conductivity is evaluated using the Boltzmann equation. A simple expression for the reduction in conductivity due to the increase of boundary scattering is presented. A comparison is made between the experimental results of indium wires and the theoretical calculations. It is shown that this decrease of conductivity in wires is smaller than that in film where heat flux is perpendicular to the surface.

Keywords: thermal conductivity, boundary scattering
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I. INTRODUCTION

Although many studies have been carried out on the thermal conductivity of thin films,[1−3] less investigation has been done on the conductivity of metallic thin wires. In this paper, the effect of radial thickness on thermal conductivity is investigated by considering free electron transport using the Boltzmann equation. The imposed temperature gradient is along the wire, as is the direction of the heat flow. We can neglect the boundary scattering of radial thickness for thick wire or bulk samples. However, the scattering becomes increasingly dominant as the radial thickness decreases. For the radial thickness of the order of mean free path (MFP), boundary scattering of carriers is expected to play a significant role. We examine the reduction in the thermal conductivity of thin metallic wire where the primary heat carriers are free electrons. Metallic wire plays an important role in applications, such as ultra-large scale integration (ULSI) and microelectromechanical system (MEMS) technology. Metallic wires are used as interconnects for microelectronics. For example, Advanced Micro Devices Incorporated (AMD) and Intel Incorporated have begun to use 0.18 μm interconnects technology in their CPU fabrication. Size effects on thermal conductivity play a vital role there.

II. THERMAL CONDUCTIVITY OF THE WIRES OF CIRCULAR CROSS SECTION

The dependence of the thermal conductivity of wire on its diameter primarily arises from their electrical conductivity, because the thermal conductivity of a pure bulk sample is proportional to its electrical conductivity according to the Wiedemann–Franz law. It is well known that electrons in metal serve as both electrical and thermal carriers. The phonon contribution to the thermal conductivity in metal is negligible. This law is expressed as

$$\frac{\kappa}{\sigma T} = \frac{\pi^2 k^2}{3e^2} = L = 2.45 \times 10^{-8} \mathrm{W\Omega/K^2}, \qquad (1)$$

where κ and σ are the thermal and electrical conductivities, L is the Lorenz number, k is the Boltzmann constant, e is the electron charge and T is the temperature. Equation (1) is theoretically and experimentally satisfied for bulk metal, but has not been proved for thin films and wires. Recently, Starz[4] and Stewart[5] have proved that Eq.(1) is also valid for thin films and wires. By using the formula, Nath[6] calculated the effective size effects for the thermal conductivity. Using the electrical–thermal transport analogy, the thermal conductivity of the metallic wire can be written as

$$\frac{\kappa_{\mathrm{wire}}}{\kappa_{\mathrm{bulk}}} = \psi\left(\frac{2a}{\lambda}, \epsilon\right) = \psi\left(\frac{d}{\lambda}, \epsilon\right), \qquad (2)$$

where κ_{wire} is the thermal conductivity of the wire, κ_{bulk} is the conductivity of the bulk, λ is the MFP of electrons for thermal conductivity, a and d are the radius and diameter of the wire respectively, ϵ is a specularity parameter. The function ψ was obtained by Sondheimer.[7] However, the thermal conductivity is determined by heat flow resulting from a temperature gradient in the condition that there is no electronic

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current. The thermal conductivity of the metallic wire is calculated using the simplified form of the steady-state Boltzmann equation. We consider a cylindrical wire of radius a, and take a temperature gradient, $\nabla T = \dfrac{\partial T}{\partial z}$, to be along the wire axis, which is defined as the z direction, as shown in Fig.1. The linear Boltzmann equation in the relaxation time approximation is

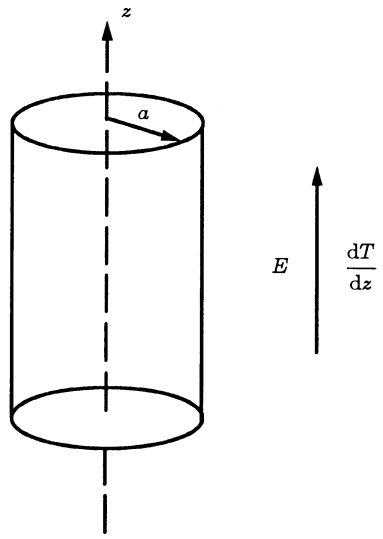


Fig.1. Schematic diagram of the system under consideration.

$$-\frac{eE_z}{m}\frac{\partial f}{\partial v_z}+v_z\frac{\partial f}{\partial T}\frac{\mathrm{d}T}{\mathrm{d}z}+v_x\frac{\partial f}{\partial x}+v_y\frac{\partial f}{\partial y}=-\frac{f-f_0}{\tau}, \quad (3)$$

where v_x, v_y and v_z are the components of $\boldsymbol{v}$, f is the number of electrons per unit volume of ordinary space. Here E_z is the electric field set up in the z direction, m is the electron mass, and τ is the effective relaxation time.

The equilibrium function f_0 is a function of kinetic energy ε and is given by Fermi–Dirac statistics

$$f_0=2\left(\frac{m}{h}\right)^3 g_0, g_0=\frac{1}{\exp\left(\dfrac{\varepsilon-\mu}{kT}\right)+1}, \varepsilon=\frac{1}{2}m\left|\boldsymbol{v}\right|^2, \quad (4)$$

where μ is the Fermi level.

The electric current density $j_z(r)$ and its average $\overline{j_z(r)}$ are given by

$$j_z(r) = -e\int v_z f(\boldsymbol{v},r)\,\mathrm{d}\boldsymbol{V},$$
$$\overline{j_z(r)} = \frac{2}{a^2}\int_0^a j_z(r)r\,\mathrm{d}r. \quad (5)$$

The heat flux $q_z(r)$ and average heat flux $\overline{q_z(r)}$ are given by

$$q_z(r) = \int \varepsilon v_z f(\boldsymbol{v},r)\,\mathrm{d}\boldsymbol{V}, \overline{q_z(r)} = \frac{2}{a^2}\int_0^a q_z(r)r\,\mathrm{d}r. \quad (6)$$

where $\mathrm{d}\boldsymbol{V} = \mathrm{d}x\mathrm{d}y\mathrm{d}z$. For a reasonable small field, $f-f_0$ will be small. Putting $f-f_0=f_1$ and neglecting the product $E_z f_1$, Eq.(3) becomes

$$-\frac{eE_z}{m}\frac{\partial f_0}{\partial v_z}+v_z\frac{\partial f_0}{\partial T}\frac{\mathrm{d}T}{\mathrm{d}z}+v_x\frac{\partial f_1}{\partial x}+v_y\frac{\partial f_1}{\partial y}=-\frac{f_1}{\tau}. \quad (7)$$

The solution of Eq.(7) is

$$\begin{aligned} f_1 &= \left[\frac{eE\tau}{m}\frac{\partial f_0}{\partial v_z}-v_z\tau\frac{\partial f_0}{\partial T}\frac{\mathrm{d}T}{\mathrm{d}z}\right] \\ &\quad\cdot\left[1-f(rv_\theta,v_r^2+v_\theta^2)\exp\left(-\frac{1}{\tau}\frac{rv_r}{v_r^2+v_\theta^2}\right)\right] \\ &= \left[\frac{eE\tau}{m}\frac{\partial f_0}{\partial v_z}-v_z\tau\frac{\partial f_0}{\partial T}\frac{\mathrm{d}T}{\mathrm{d}z}\right]\left[1-(1-\epsilon)\sum_{j=0}^{\infty}\right. \\ &\quad\left.\cdot\epsilon^j\exp\left\{-\frac{r\sin\phi+(2j+1)\sqrt{a^2-r^2\cos\phi}}{\tau j\sin\theta}\right\}\right], \end{aligned} \quad (8)$$

with $0<\theta<\pi$ and $0<\phi<2\pi$. The element of volume in velocity space is $v^2\sin\theta\mathrm{d}v\mathrm{d}\theta\mathrm{d}\phi$. After integration over the angles, an integral over r is given to yield the average current and heat fluxes. The current is given by

$$\overline{j_z(r)} = \frac{CAe\Theta}{T}\left[\left\{eE+T\frac{\mathrm{d}}{\mathrm{d}T}\left(\frac{\mu}{T}\right)\frac{\mathrm{d}T}{\mathrm{d}z}\right\}I_{p_3} + \frac{1}{T}\frac{\mathrm{d}T}{\mathrm{d}z}I_{p_4}\right], \quad (9)$$

where $A=\dfrac{16}{3}\sqrt{2}\pi e\dfrac{\sqrt{m}}{h^3}$, Θ is the Debye temperature and C is a constant that is related to the properties of the material. I_n is defined by

$$I_n = -\int_0^\infty \varepsilon^n\frac{\partial g_0}{\partial\varepsilon}\mathrm{d}\varepsilon. \quad (10)$$

When we set the current to zero, we get

$$\kappa_{\mathrm{wire}} = \frac{CA\Theta}{T^2}\frac{I_{p_3}I_{p_5}-I_{p_4}^2}{I_{p_3}}, \quad (11)$$

$$\kappa_{\mathrm{bulk}} = \frac{CA\Theta}{T^2}\frac{I_3I_5-I_4^2}{I_3}. \quad (12)$$

I_{p_n} is defined by

$$I_{p_n} = -\int_0^\infty \varepsilon^n\beta_p(\varepsilon)\frac{\partial g_0}{\partial\varepsilon}\mathrm{d}\varepsilon, \quad (13)$$

$$\beta_p = 1-\frac{12}{\pi}(1-\epsilon)^2\sum_{j=1}^{\infty}j\epsilon^{j-1} \cdot\int_0^1\sqrt{1-x^2}S_4(j\gamma x)\,\mathrm{d}x, \quad (14)$$

with

$$S_n(u) = \int_0^{\frac{\pi}{2}} \exp\left(-\frac{u}{\sin\theta}\right) \cos^2\theta \sin^{n-3}\theta \, \mathrm{d}\theta. \quad (15)$$

where $\gamma = d/\lambda$ is the thickness ratio. When we set $\mathrm{d}T/\mathrm{d}z$ to zero, we get

$$\sigma_{\text{wire}} = \frac{CAe^2\Theta}{T} I_{p_3}, \quad (16)$$

$$\sigma_{\text{bulk}} = \frac{CAe^2\Theta}{T} I_3, \quad (17)$$

then

$$\frac{\kappa_{\text{wire}}}{\sigma_{\text{wire}} T} = \frac{1}{e^2T^2} \frac{I_{p_3} I_{p_5} - I_{p_4}^2}{I_{p_3}^2}. \quad (18)$$

It is shown that the Lorenz number is not a constant and it is related to the integral I_{p_n}. Now let us discuss a simple case: for bulk sample

$$\frac{\kappa_{\text{bulk}}}{\sigma_{\text{bulk}} T} = \frac{1}{e^2T^2} \frac{I_3 I_5 - I_4^2}{I_3^2} \approx \frac{1}{e^2} \left[\frac{\pi^2 k^2}{3B} + \frac{\pi^4 k^4 \mu^2 T^2}{15B}\right], \quad (19)$$

where $B = 1 + \pi^2(kT/\mu)^2$. Because $kT \ll \mu$ at room temperature, we have $B \approx 1$ in this situation. Equation (19) yields

$$\frac{\kappa_{\text{bulk}}}{\sigma_{\text{bulk}} T} = \frac{\pi^2 k^2}{3e^2}. \quad (20)$$

This is the same as the well-known Wiedemann–Franz law. We have shown that this law is valid for a bulk sample. Now we use asymptotic expansion[8] to evaluate the ratio of thermal conductivity of the wire to that of the bulk,

$$\frac{\kappa_{\text{wire}}}{\kappa_{\text{bulk}}} \approx 1 - \frac{12}{\pi}(1-\epsilon)^2 \sum_{j=1}^{\infty} j\epsilon^{j-1} \cdot \int_0^1 \sqrt{1-x^2} S_4(j\gamma x)\, \mathrm{d}x, \quad (21)$$

with

$$\kappa_{\text{bulk}} \approx \frac{\pi^2 n k^2 T \tau}{3m}, \quad (22)$$

where n is the density of the carriers. This is the same result as the thermal conductivity under the free electron approximation. The results about the ratios the conductivity of metallic wires to that of bulk material are the same as the expression obtained by Dingle[9] for the electrical conductivity ratio. This indicates that the ratio of the thermal conductivity of metallic thin wire to that of the bulk material is identical with that for electrical conductivity of thin wire.

III. THERMAL CONDUCTIVITY WITH BOUNDARY SCATTERING

Now we discuss the thermal conductivity with boundary scattering. Figure 2 shows the curves of the thermal conductivity ratio versus thickness ratio γ for different values of the specularity scattering fraction ϵ in Eq.(21). If all the electrons are scattered (ϵ=1), then the wire conductivity equals the bulk conductivity. Figure 3 shows the wire conductivity ratio versus the specularity scattering fraction ϵ and thickness ratio γ. We can find that the thermal conductivity increases monotonically with increasing ϵ or γ; ϵ is a parameter related to the amount of the specularity scattering electrons. So the more the specularity scattering electrons, the greater the heat conduction, and the larger the thermal conductivity. When γ becomes large, the carriers have the chance to go farther before they are diffused. This leads to larger thermal conductivity. Equation (21) is difficult to evaluate directly, so we derive the following approximations by expanding the root in Eq.(21)

$$\frac{\kappa_{\text{wire}}}{\kappa_{\text{bulk}}} \approx \begin{cases} 1 - \dfrac{3}{4\gamma}(1-\epsilon) + \dfrac{3}{8\gamma^3}(1-\epsilon)^2 \displaystyle\sum_{j=1}^{\infty} \epsilon^{j-1}/j^2 \\ (\gamma \gg 1), \\ \dfrac{1+\epsilon}{1-\epsilon}\gamma - \dfrac{3}{8}\Big[\dfrac{\epsilon^2+4\epsilon+1}{(1-\epsilon)^2}\{\ln(1/\gamma)+1\} \\ -(1-\epsilon)^2 \displaystyle\sum_{j=1}^{\infty} j^3\epsilon^{j-1}\ln j\Big]\gamma^2 \quad (\gamma \ll 1). \end{cases} \quad (23)$$

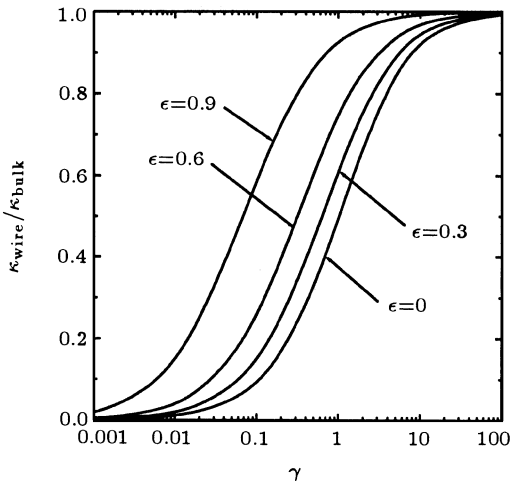


Fig.2. The thermal conductivity ratio of the wire to the bulk material, as a function of thickness ratio γ for different values of the specularity scattering fraction ϵ.

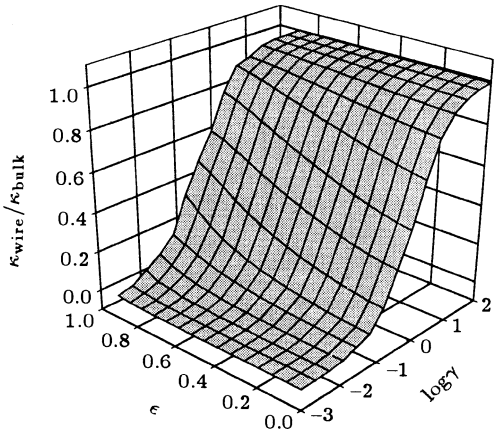


Fig.3. The thermal conductivity ratio of the wire to the bulk material versus γ and the specularity scattering fraction ϵ.

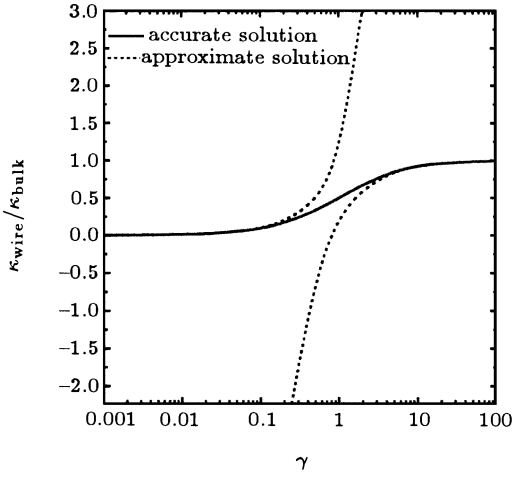


Fig.4. The approximate and the accurate solutions of Eq.(21).

For $\epsilon = 0$, Eq.(23) reduces to

$$\frac{\kappa_{\text{wire}}}{\kappa_{\text{bulk}}} \approx \begin{cases} 1 - 3/(4\gamma) & (\gamma \gg 1) \\ \gamma - \dfrac{3}{8}\ln(1/\gamma) & (\gamma \ll 1) \end{cases} \tag{24}$$

The above expressions are compared with Eq.(21) for ϵ=0 in Fig.4. We can find that the approximate expressions are accurate for large γ or small γ. The experimental data from Ref.[10] for indium wires are shown in Fig.5. It shows the value of thermal conductivity ratio plotted against wire diameter ratio (ratio of diameter to electron MFP). We note that these thermal conductivity ratios can also be used to represent the electrical conductivity ratio of the material.

Now we use some models to fit the experimental data of thermal conductivity ratio of indium wires. At first we use the modified relaxation time model. Bate[10] thought that only lines drawn nearly parallel to the axis will intersect a large number of grain boundaries in the wire. If the grain boundary scattering is considered to be modifier of the relaxation time τ in the wire, while the grain size will increase with the thickness or diameter, we can get the thermal conductivity ratios by Matthiessen's rule

$$\frac{\kappa_{\text{wire}}}{\kappa_{\text{bulk}}} = \frac{1}{1+D/\gamma}\left\{1 - \frac{12}{\pi}(1-\epsilon)^2 \sum_{j=1}^{\infty} j\epsilon^{j-1} \cdot \int_0^1 \sqrt{1-x^2}\, S_4\left[j\gamma\left(1+\frac{D}{\gamma}\right)x\right] \mathrm{d}x\right\}, \tag{25}$$

with

$$D = \frac{d}{d_g}, \tag{26}$$

where d_g is the average in-plane grain diameter. It is considered to be proportional to the wire radial thickness. D is the proportionality constant. The result of Eq.(25) is plotted in Fig.5 along with the experimental data. When $\gamma + D \gg 1$, expanding the root in Eq.(25) and integrating over x, we obtain

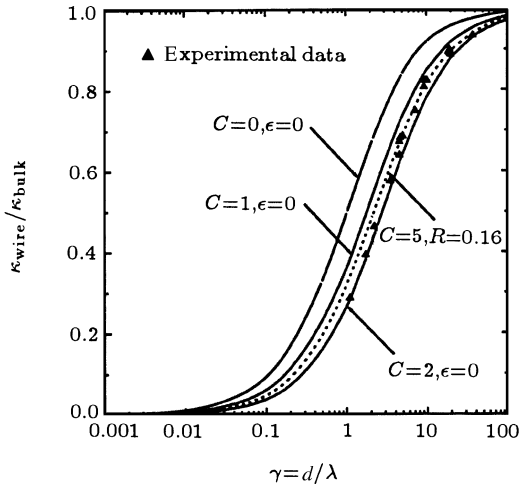


Fig.5. The thermal conductivity ratio of the wire to the bulk material for diffusive scattering surface with grain boundaries.

$$\frac{\kappa_{\text{wire}}}{\kappa_{\text{bulk}}} \approx \frac{1}{1+D/\gamma}\left[1 - \frac{3}{4}\frac{1}{\gamma+D} + \frac{3}{8}\frac{1}{(\gamma+D)^3}\right]. \tag{27}$$

Another model is Qiu and Tien's model.[11] They combined both the surface and grain boundary scattering effects. However, they only evaluated the reduction in thermal conductivity of thin films when heat flux is parallel to the surface. Now we generalize it to thin metallic wires and thin films when the heat flux is perpendicular to the surface. De Vries[12] evaluated the associated reduction in the thermal conductivity

$$\frac{\kappa}{\kappa_{\text{bulk}}} = 1 - \frac{3}{2}\alpha + 3\alpha^2 - 3\alpha^3 \ln(1+1/\alpha), \tag{28}$$

with

$$\alpha = \frac{D}{\gamma}\frac{R}{1-R}. \tag{29}$$

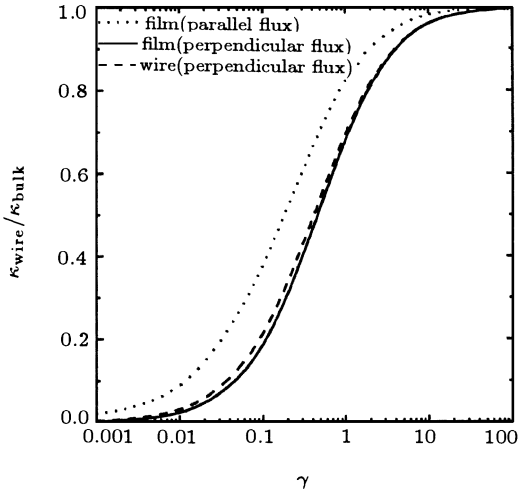


Fig.6. The conductivity ratios of the film to the bulk material, and of the wire to the bulk material, as a function of γ (ϵ=0.5).

Here R is the reflection coefficient of the conduction electrons striking the grain boundaries ($0 \le R \le 1$). Then we combine the two scattering mechanisms to get the thermal conductivity ratio for $\epsilon = 0$,

$$\frac{\kappa_{\rm wire}}{\kappa_{\rm bulk}} = \frac{\kappa_g}{\kappa_{\rm bulk}} - \frac{12}{\pi\gamma}\int_1^\infty {\rm d}tH^{-2}(t)\frac{1}{t^4}\sqrt{1-\frac{1}{t^2}} \cdot \left[1 - \int_0^1 \exp\left(-\frac{\gamma H(t)u}{\sqrt{1-\frac{1}{t^2}}}\right)\frac{u}{\sqrt{1-u^2}}{\rm d}u\right], \tag{30}$$

$$\frac{\kappa_{\rm film\perp}}{\kappa_{\rm bulk}} = \frac{\kappa_g}{\kappa_{\rm bulk}} - \frac{3}{\gamma}\int_1^\infty {\rm d}tH^{-2}(t) \cdot \frac{1}{t^5}[1-\exp(-\gamma H(t)t)], \tag{31}$$

with

$$\kappa_g = 3\kappa_{\rm bulk}\int_0^{\pi/2} {\rm d}\theta \frac{\sin\theta\cos^2\theta}{H(\theta)}, \tag{32}$$

where $H(\theta) = 1 + \alpha/\cos\theta$ and $t = 1/\cos\theta$. The result of Eq.(30) is also plotted in Fig.5 along with the experimental data. We use the relationship between d and d_g. De Vries suggested D is about 5. Using Eq.(30) with R=0.16 to fit the data in Fig.5, it is shown that our approximation model agrees better with the data than Eq.(25). The results show that the thermal conductivity values of indium can be explained by our combination model.

We have calculated the thermal conductivity of metallic wires, the result of which is compared with that of thin films, as shown in Fig.6. It shows $\kappa_{\rm wire}$ and κ of the film in which heat flux is either parallel or perpendicular to the surface. They are scaled by the bulk value $\kappa_{\rm bulk}$, for partly diffusive scattering (ϵ=0.5) and for γ ranging between 0 and 100. It is shown that $\kappa_{\rm wire}$ and $\kappa_{\rm film}$ increase below the bulk value with increasing film and wire thickness. This is a consequence of the decrease of boundary scattering that occurs as the film and wire thickness increases. We can get the limiting form of the scaled thermal conductivity for $\gamma \gg 1$ and $\gamma \ll 1$

$$\frac{\kappa_{\rm film\parallel}}{\kappa_{\rm bulk}} \approx \begin{cases} 1 - \dfrac{3}{8\gamma}(1-\epsilon) & (\gamma \gg 1), \\ 1-(1-\epsilon)^2\displaystyle\sum_{j=1}^{\infty} j\epsilon^{j-1} & \\ \cdot\left\{1 - \dfrac{3}{4}j\gamma\left[1 - \ln(j\gamma) - c + \dfrac{2}{3}j\gamma\right]\right\} & (\gamma \ll 1), \end{cases} \tag{33}$$

$$\frac{\kappa_{\rm film\perp}}{\kappa_{\rm bulk}} \approx \begin{cases} 1 - \dfrac{3}{4\gamma}(1-\epsilon) & (\gamma \gg 1), \\ 1-(1-\epsilon)^2\displaystyle\sum_{j=1}^{\infty} j\epsilon^{j-1}\left\{1 - \frac{3}{4}j\gamma\right. & \\ \left. + \dfrac{1}{2}j^2\gamma^2 - \dfrac{1}{8}j^3\gamma^3[1.256 - \ln(j\gamma)]\right\} & (\gamma \ll 1), \end{cases} \tag{34}$$

where c is Euler's constant used in the expansion of the exponential integral function. We find the expression of $\kappa_{\rm film\perp}$ is similar to $\kappa_{\rm wire}$ when $\gamma \gg 1$. For thick films and wires, they are like bulk materials, so that they have same thermal conductivity. The cross section of these is large, the upper and lower surfaces of films dominate the bulk scattering process. For bulk materials, it is difficult to observe size effect because

the MFP of carriers are always much shorter than the sample size at room temperature. However, for very thin samples, the MFP of carriers are longer than the size of sample even at room temperature. So it is easy to observe size effect in thin samples.

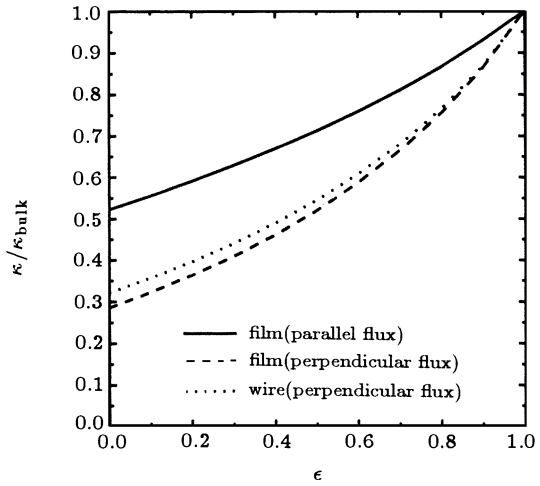


Fig.7. The conductivity ratios of the film to the bulk material, and of the wire to the bulk material, as a function ϵ (γ=0.5).

Figure 7 shows the thermal conductivity ratios as functions of ϵ for γ=0.5. When $\epsilon \to 1$, the thermal conductivities of wires and films increase up to the bulk value because the boundary scattering no longer influences the conduction. Also from Fig.7, we find that the thermal conductivity of wires at certain ϵ and γ, is smaller than that of corresponding film in which heat flux is parallel to the surface, but larger than that of the corresponding film in which heat flux is perpendicular to the surface. For the same volumes, the surface area of wires is larger than that of films, so boundary scattering is enhanced. Therefore, the thermal conductivity of wires is smaller than that of films. It is obvious that the intensity of boundary scattering is determined by the scattering component normal to the surface. When the heat flux is perpendicular to the surface, boundary scattering enhances so that the thermal conductivity reduces. This factor even exceeds the influence of surface area. Though the surface area of wires is larger than that of films of the same volume, the boundary scattering of wires is weaker than that of films in which heat flux is perpendicular to the surface. This leads to a smaller thermal conductivity of thin films.

IV. CONCLUSION

A solution of the Boltzmann transport equation is presented for metallic free standing wires. The expressions obtained predict the dramatic reduction in thermal conductivity with decreasing size because of the enhanced boundary scattering. At first grain boundary scattering is considered by the modified relaxation time model. Then we generalize Qiu and Tien's model to study wires and films in which heat flux is perpendicular to the surface. The numerical results of these models are compared with experimental data. We find that the data of indium wires agree with our model much better than the modified relaxation time model. Finally we compare the thermal conductivities of films and wires with different thicknesses. It is shown that the thermal conductivity of wires is smaller than corresponding films in which heat flux is parallel to the surface, but larger than that of the corresponding film in which heat flux is perpendicular to the surface.

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