Effective carrier mean-free path in confined geometries

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The concept of exchange length is used to determine the effects of boundary scattering on transport in samples of circular and rectangular cross section. Analytical expressions are presented for an effective mean free path for transport in the axial direction. The relationship to the phonon thermal conductivity is discussed.

The properties of transport in confined geometries has received substantial attention in recent years (see, e.g., Ref. 1). In this letter, the effects of boundary scattering on transport in small samples is examined. Our goal is to provide concise expressions for the effect of boundaries on transport without explicitly evaluating the Boltzmann equation. We present analytical expressions for the effective mean free path in samples where the bulk mean free path is determined by other scatterers present in the sample. Expressions are obtained for samples of circular and rectangular cross section. These results are applicable to samples which are small enough that the carrier mean free path is on the order of the sample dimensions but not so small that the carrier spectrum is substantially modified from the bulk. In other words, the sample dimensions will be assumed to be much greater than the carrier wavelength.

We employ a method first proposed by Flik and Tien[2] for the calculation of the size effect in thin films. The method assumes that, for a carrier of a given frequency in a bulk sample, a characteristic mean free path, l, can be defined. The goal, then, is to examine how this bulk value of l is modified by the presence of boundaries in the sample. The calculation utilizes the concept of the exchange length l_{ex},[2,3] which is defined as the average distance normal to a plane that a carrier travels after having been scattered within that plane. Specifically, we consider a carrier that has undergone a scattering event within a plane that is perpendicular to the direction of net transport, which will be referred to from here on as the positive z direction. We now allow the carrier to propagate to the point of its next scattering event, which, in the bulk, is a distance l away. This propagation is assumed to proceed with equal likelihood in all directions. l_{ex} is then defined as the average z component of all possible such propagation vectors, where the average is performed over the hemisphere in the positive z direction. The bulk value of this quantity, l_∞ is $l/2$.

In the following, we consider the scenario in which the mean free path is on the order of the sample dimensions. For this case, some carriers will strike the boundaries before traveling a full distance l and the exchange length will be correspondingly shorter. We will assume that scattering at the boundaries is diffuse, which will be valid when the carrier wavelength is smaller than the characteristic roughness features of the sample surface. This hypothesis needs to be examined within the context of a particular measurement, but holds for many materials. We note that a principle purpose of this work is to extend the most widely used form for the effects of boundary scattering on the thermal conductivity[4] and in this form, perfect sample roughness is also assumed. We will also consider the sample to be free of grain boundaries, though the expression derived could perhaps also be applied to samples whose grains have characteristic geometries which match those investigated here.

We first calculate the exchange length for axial transport in a cylindrical sample of infinite length. We initially assume that the excitation can originate with equal likelihood anywhere within a given circular cross section of the sample. The average value of the exchange length in the sample, $\tilde{l}_{ex}$, is then obtained by averaging l_{ex} (which is itself an average over a hemisphere of solid angle) over the entire cross section. The geometry to be considered is shown in Fig. 1. We consider an excitation originating at some point a distance ρ from the center of the cross section of radius R and propagating in a random direction within the hemisphere of solid angle whose base is normal to the positive z direction. The quantity θ is defined as the angle between the propagation vector l and the z axis, and ϕ is the angle between the radius along which the origination point is located and the projection of l into the plane of the cross section. Note that l may or may not have length l, depending upon whether or not it is truncated by a boundary. The average exchange length is then given by

$$\tilde{l}_{ex}=\frac{2}{\pi R^2}\int_0^R d\rho\,\rho\int_0^\pi d\phi\int_0^{\pi/2} d\theta\,\sin\theta\, l_z(\rho,\phi,\theta), \quad (1)$$

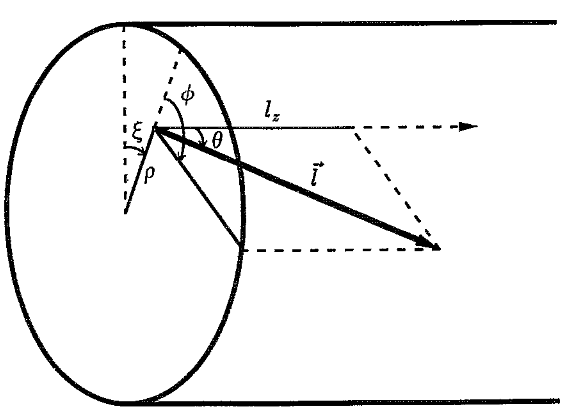

FIG. 1. Schematic diagram showing the geometry relevant for the calculation of the exchange length for a wire of circular cross section. The case pictured is that for which l hits the boundary.

TABLE I. Summary of the equations for $\tilde{l}_{ex}$ for the various different geometries and the various ranges of the mean free path l. R is the radius for the circular case, and a and b are the lengths of the sides for the rectangular case, with b taken to be the shorter of the two. d is equal to $\sqrt{a^2+b^2}$.

Cross section	Range	$\tilde{l}_{ex}$
Circle	$l<2R$	$\frac{8R}{3\pi}-\frac{R^2}{2l}+\frac{l}{\pi}\cos^{-1}\left(\frac{l}{2R}\right)+\frac{R^2}{2\pi l}\cos^{-1}\left(\frac{l^2}{2R^2}-1\right)-\frac{(4R^2-l^2)^{1/2}}{\pi}\left(\frac{13}{12}+\frac{l^2}{24R^2}\right)$
	$l\geqslant 2R$	$\frac{8R}{3\pi}-\frac{R^2}{2l}+\exp(-8R/l)\left(\frac{4R}{3\pi}-\frac{R^2}{2l}\right)$
Rectangle	$l<b$	$F(a,b)$
	$b\leqslant l<a$	$F(a,b)+G(b,a)$
	$a\leqslant l<d$	$F(a,b)+G(a,b)+G(b,a)$
	$l\geqslant d$	$H+\exp(-4d/l)(J-H)$
Square	$l<a$	$F(a,a)$
	$a\leqslant l<\sqrt{2}a$	$F(a,a)+2G(a,a)$
	$l\geqslant\sqrt{2}a$	$\frac{a}{\pi}\left[2+\exp\left(-\frac{4\sqrt{2}a}{l}\right)\right][\ln(1+\sqrt{2})+(1-\sqrt{2})/3]-\frac{a^2}{2\pi l}\left[1+\exp\left(-\frac{4\sqrt{2}a}{l}\right)\right]$

$$F(p,q)\equiv\frac{l}{2}-\frac{(p+q)}{3\pi pq}l^2+\frac{l^3}{12\pi pq};\quad G(p,q)\equiv-\frac{(p-l)^4}{12\pi pql}+\frac{p}{\pi}\ln\left(\frac{l}{p}+\sqrt{(l/p)^2-1}\right)-\frac{l}{\pi}\cos^{-1}\left(\frac{p}{l}\right)+\frac{(l^2-p^2)^{3/2}}{3\pi pl};$$

$$H\equiv\frac{a}{\pi}\ln\left(\frac{b+d}{a}\right)+\frac{b}{\pi}\ln\left(\frac{a+d}{b}\right)+\frac{1}{3\pi ab}(a^3+b^3-d^3)-\frac{ab}{2\pi l};\quad J\equiv\frac{2}{\pi(a+b)}\left[\left(\frac{a^2}{2}+ab\right)\ln\left(\frac{b+d}{a}\right)+\left(\frac{b^2}{2}+ab\right)\ln\left(\frac{a+d}{b}\right)\right]+\frac{d^2}{\pi(a+b)}-\frac{d}{\pi}-\frac{ab}{\pi l}$$

where l_z is the z component of the propagation vector heading in the (θ,ϕ) direction and ϕ has only been integrated over half its range for symmetry reasons.

The evaluation of this integral involves a careful analysis of the regions of (ρ,ϕ,θ) space where l does and does not hit the wall. This is of importance since the functional form of l_z clearly depends on whether or not l is truncated by a collision with a wall. The details of this process are beyond the scope of this article and will be presented in a more comprehensive work.[5] The expressions for $\tilde{l}_{ex}$ for the circular case are given in Table I.

We have also evaluated the average exchange length for samples of rectangular cross section. The general expression for $\tilde{l}_{ex}$ is quite analogous to the circular case.[5]

As in the circular case, the evaluation of $\tilde{l}_{ex}$ depends upon discerning which regions of phase space have $\tilde{l}_{ex}$, hitting the wall and which do not. The analysis of this problem will be presented in detail in Ref. 5. Because of the reduced symmetry of this case relative to the circular one, the solution needs to be broken down into more regimes. These results are shown in Table I.

The expressions presented thus far for the evaluation of $\tilde{l}_{ex}$, for both the circular and rectangular cases, were derived on the assumption of uniform origination, i.e., we assume that the excitation can originate anywhere within the cross section with equal likelihood. As pointed out in Ref. 2, however, when the mean-free path of the excitation becomes much longer than the sample dimensions, the excitation becomes increasingly likely to scatter on a boundary and our assumption of uniform origination needs to be replaced with a boundary origination description. The calculation of the boundary origination solutions is straightforward and the expressions for the two geometries are as follows: Circular: $\tilde{l}_{ex}=(4R/\pi)-(R^2/l)$; rectangular: $\tilde{l}_{ex}=J$, where J is given in Table I.

The appropriate procedure[2] is to match these solutions to those for uniform origination at large l's. This can be accomplished by a simple exponential matching process, whereby the matched solution is obtained by adding the uniform origination solution to the difference between the boundary and uniform solutions times an exponential function. This process results in the equations in Table I where $l\geqslant 2R$ for the circular case and $l\geqslant d$ for the rectangular case.

These forms result in small discontinuities when they are combined with the solutions for $l<2R$ and $l<d$, respectively. The matched solutions contain a matching parameter in the exponential functions which we choose to be four to achieve the best compromise between minimizing this discontinuity and "phasing in" the boundary origination solution as quickly as possible as l increases. It should be noted that, for the rectangular case when $a\gg b$, we can expect to be largely in the boundary origination regime before l exceeds the length of the diagonal. The above equation does not allow for this possibility and is therefore most applicable to cases in which a is not too different from b. For samples where one dimension is much larger than the other, we refer the reader to the results of Ref. 2 where an expression for thin films is derived.

The expressions given in Table I can now be used to aid in the calculation of transport quantities in samples where boundary scattering is expected to play a role. The general procedure is as follows. We know that the bulk mean free path is given by two times the the bulk exchange length. Now, this relationship can be extended to the confined geometry case by defining an effective mean free path for axial transport as $l_{eff}=2\tilde{l}_{ex}$, where $\tilde{l}_{ex}$ for axial transport is given by the expressions in Table I.

Let us apply this relationship to the thermal conductivity. In the kinetic theory approximation, the thermal conductivity κ is given by $\kappa = \frac{1}{3}Cvl$, where C is the contribution to the specific heat from the carrier in question and v is the carrier velocity. Now, in Ref. 2, it is asserted that, within this approximation, the transport κ_z along the z axis in a sample of confined geometry can be obtained from the bulk thermal conductivity κ_∞ and the exchange length along that axis by the expression $\kappa_z = \kappa_\infty \tilde{l}_{ex}/(l/2)$. It can easily be seen then that the value of the thermal conductivity for the confined sample is obtained from the bulk expression by simply substituting l_{eff} for l, i.e., $\kappa_z(l) = \kappa_\infty(l_{eff})$, is defined above.

Because of the simple geometric nature of this argument, it is plausible that this sort of analysis can be applied to more sophisticated treatments of the thermal conductivity as well. For instance, in the case of phonon transport, the thermal conductivity is often written in the Debye approximation as an integral over phonon frequencies:[6]

$$\kappa_p(T) = \frac{k_B}{2\pi^2 v}\left(\frac{k_B}{\hbar}\right)^3 T^3 \int_0^{\theta_D/T} dx \frac{x^4 e^x}{(e^x-1)^2}\tau(T,x), \quad (2)$$

where x is the reduced phonon frequency $\hbar\omega/k_B T$, k_B is the Boltzmann constant, θ_D is the Debye temperature, and $\tau(T,x)$ is the frequency dependent scattering time. The total inverse scattering time, $\tau(T,x)^{-1}$, is usually expressed as a summation of the inverse scattering times from scatterers of various types. Within this context, the effect of boundaries is typically handled by a method due to Casimir[4] whereby one adds a frequency independent term to this total of the form $\tau_b^{-1} = v/\alpha d$, where d represents the sample dimension and α is a geometrical factor.

We propose that greater accuracy may be achieved from calculations involving Eq. (2) by omitting the boundary scattering term in the total inverse scattering time and utilizing the equations in Table I for the exchange length to modify the mean free path instead. The proposed procedure is as follows. The factor τ in Eq. (2) can readily be replaced by $l(x,T)/v$, at which point the integration can be seen to be over the frequency-dependent mean free path multiplied by another x dependent factor. For each such mean free path, $l(x,T)$, a value of $\tilde{l}_{ex}$ can be derived by using the equation appropriate for the geometry of the particular sample under investigation. Each $l(x,T)$ in the integral can then be replaced by an $l_{eff}(x,T)$ as described above. The proper boundary limited value of the thermal conductivity is then obtained by integrating over the $l_{eff}(x,T)$, with the other factors in Eq. (2) left unaltered.

An example of this process is pictured in Fig. 2. The graph shows two calculations of the thermal conductivity of diamond using Eq. (2) as a basis. The solid curve is taken from a recent work[7] and uses the simple treatment whereby the boundary scattering is handled by the addition of a constant term to the inverse scattering time.[4] The form of the constant term is for axial transport along perfectly rough grains of square cross section ($\alpha = 1.12$). The model also includes a point defect scattering term and a phonon–phonon umklapp term. The dashed curve shows the results when all of the parameters of the model are left

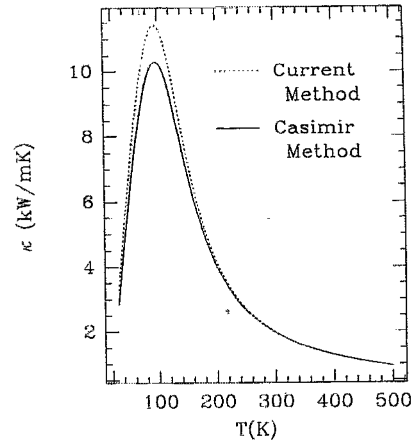

FIG. 2. Calculation of the thermal conductivity of diamond using the current and Casimir methods. The present technique produces a 12% enhancement in the peak relative to the Casimir method. Model parameters are taken from Ref. 7.

unchanged but the boundary scattering is treated by the method described in this work (the square cross section equations are used). The assumptions about direction of transport and surface roughness are the same as for the solid curve. One can see that the present method produces a significant enhancement of the thermal conductivity peak relative to the Casimir method.

In summary, we have presented analytical expressions for the effects of boundary scattering in samples where the bulk carrier mean free path is determined by other scatterers present. Results are derived for axial transport in long, narrow samples of circular and rectangular cross section, where scattering at the boundaries is diffuse. The results are incorporated into a definition of an effective mean free path for axial transport which can be used to calculate coefficients such as the thermal conductivity. Though we have focused on thermal transport in the present work, the expressions derived here could be of use in the examination of a variety of transport phenomena in confined geometries.

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