

Thermal Conduction in Nonhomogeneous CVD Diamond Layers in Electronic Microstructures

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Chemical-vapor-deposited diamond layers of thickness between 0.1 and 5 μm have the potential to improve conduction cooling in electronic microstructures. However, thermal conduction in these layers is strongly impeded by phonon scattering on defects, whose concentrations can be highly nonhomogeneous, and on layer boundaries. By assuming that defects are concentrated near grain boundaries, this work relates the internal phonon scattering rate to the local characteristic grain dimension and to the dimensionless grain-boundary scattering strength, a parameter defined here that varies little within a given layer. Solutions to the Peierls-Boltzmann phonon transport equation for conduction along and normal to layers account for the nonhomogeneous internal scattering rate. Predictions for conduction along and normal to layers as thin as 0.2 μm agree well with room-temperature data. This research helps optimize diamond layer thicknesses for specific microstructures, such as silicon-on-diamond (SOD) circuits.

1 Introduction

At room temperature, chemical-vapor-deposited (CVD) diamond can be both an excellent thermal conductor and an electrical insulator, making it ideal for passive use in electronic microstructures. The benefit is greatest if diamond can be brought within micrometers of active regions, typically made of silicon or gallium arsenide (Goodson, 1995). The small separation allows the diamond to reduce the problem of local hotspots with micrometer-scale dimensions, which degrade the performance and reliability of electronic systems. One example is a high-power-density laser-diode array based on a novel composite diamond/gallium-arsenide substrate (Goodson et al., 1995c). Another example is a silicon-on-diamond (SOD) circuit in which a thin diamond layer separates active silicon regions from the substrate (Ravi and Landstrass, 1989). SOD circuits are a novel form of silicon-on-insulator (SOI) technology, which improve the speed and voltage-blocking capability of devices through the use of a buried insulating layer. The use of diamond as the buried insulating layer offers the promise of avoiding the thermal resistance due to conventional buried insulators, such as silicon dioxide (Goodson et al., 1995a). Temperature fields in SOD circuits are strongly influenced by conduction along and normal to diamond layers thinner than 5 μm .

The deposition of thin diamond layers on silicon yields a highly imperfect and nonhomogeneous material, particularly near the interface. This is evident in the grain structure, which is shown in the cross-sectional micrograph in Fig. 1 and approximately depicted in Fig. 2. The nucleation of diamond on a silicon substrate results in very small grains near the interface (e.g., Stoner et al., 1992). The scattering rate of phonons, which are responsible for heat conduction in diamond, is strongly influenced by the local grain size. However, the relationship between the phonon scattering rate and the grain size has not

been rigorously determined. In thin layers, heat conduction is influenced by the strong nonhomogeneity of the phonon scattering rate, which results from the varying grain size, and by phonon scattering on the layer boundaries. Predictions require solutions to the phonon transport equation in nonhomogeneous layers, which are not available.

Previous data demonstrated the strong dependence of the local thermal conductivity along diamond layers on the coordinate z in Fig. 2 (Graebner et al., 1992a, b; Käding et al., 1993, 1994). The room-temperature conductivity varies from approximately 2000 $\text{W m}^{-1} \text{K}^{-1}$ for values of z greater than about 150 μm , to values below the conductivity of silicon, 150 $\text{W m}^{-1} \text{K}^{-1}$, for values of z less than about 5 μm . Recent work measured the effective thermal conductivities normal to thinner layers, obtaining values between 12 and 75 $\text{W m}^{-1} \text{K}^{-1}$ for layers of thickness 0.2 and 2.6 μm , respectively (Goodson et al., 1995b). Previous analysis solved the transient equation of phonon radiative transfer for conduction normal to diamond layers using the homogeneous phonon-scattering parameters of nearly perfect crystals (Joshi and Majumdar, 1993). There is a need to solve the transport equation considering the nonhomogeneous, highly imperfect conditions in contemporary layers of thickness less than a few micrometers.

This work develops a theory for the dependence of the local phonon scattering rate in diamond layers on the local characteristic dimension of grains, d_G . The phonon transport equation is integrated for the case of conduction along a layer with a nonhomogeneous phonon mean free path, yielding the effective thermal conductivity as a function of layer thickness, temperature, and parameters describing the grain structure. An approximate solution to the transport equation for the case of conduction normal to a nonhomogeneous layer is also given. Predictions for conduction along and normal to diamond layers as thin as 0.2 μm are compared with experimental data. This work is of interest to researchers fitting the temperature-dependent conductivity in diamond polycrystals, who lack a precise treatment of the scattering rate due to grain boundaries. This research helps quantify the benefit of using passive diamond layers in electronic microstructures, which is governed by the ability of the layers to conduct heat from active semiconducting regions.

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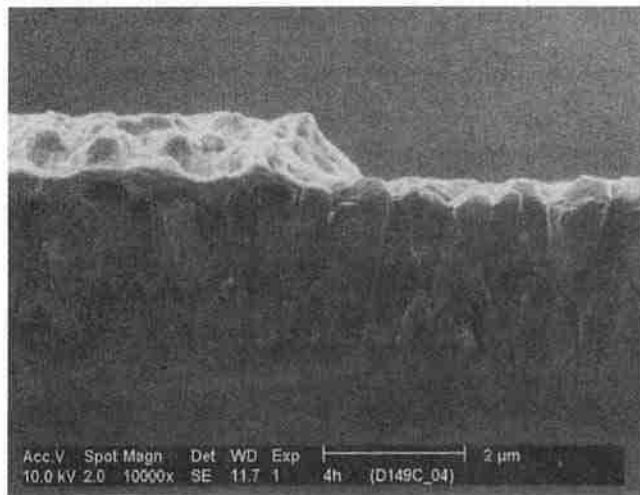


Fig. 1 Cross-sectional scanning electron microscopy image of a partially metallized diamond layer on silicon. The diamond layer thickness is $2.6 \mu\text{m}$ and the metallization is gold with a titanium adhesion layer. This structure was used by Goodson et al. (1995b) to measure the thermal resistance for conduction normal to the diamond layer into the silicon.

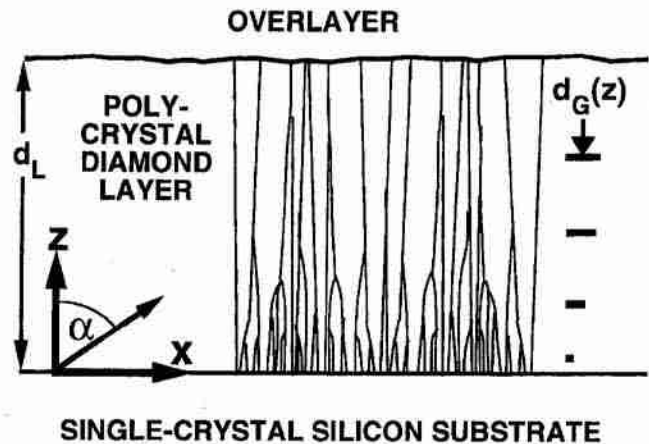


Fig. 2 Grain structure in CVD diamond layers

2 Grain Boundaries and the Local Phonon Scattering Rate

Phonon thermal conduction in diamond layers can be described using a phonon relaxation time, τ (e.g., Graebner, 1993). The total phonon scattering rate, $1/\tau$, is given by

$$[\tau(z, \omega, T)]^{-1} = [\tau_U(\omega, T)]^{-1} + [\tau_D(z, \omega)]^{-1} \quad (1)$$

The terms in Eq. (1) depend on the phonon angular frequency, ω , the temperature, T , and the position in the layer with respect to the substrate on which it was deposited, z . The first term on

the right is due to phonon-phonon Umklapp scattering. The second term on the right is the phonon scattering rate due to defects, which is strongly influenced by the grain structure. The grain structure is described by the characteristic grain dimension, $d_G(z)$, which is defined as the average distance between intersections of grain boundaries with a reference line placed on a top-view electron micrograph of a layer of thickness z .

Scattering on diamond grain boundaries is at the root of a puzzle. Diamond layers with columnar grains have higher room-temperature thermal conductivities in the z direction, i.e., parallel to grain boundaries, than in the x direction (e.g., Graebner, 1993), suggesting that scattering on grain boundaries is important. But the importance of scattering on grain boundaries is questionable, due to the lack of an appropriate signature in the measured temperature dependence of the low-temperature thermal conductivity (e.g., Morelli et al., 1993). At tempera-

Nomenclature

A_U = fitting parameter, relaxation time for scattering on phonons, $s^{-1} K^{-3}$	N_0 = equilibrium phonon distribution function	β = angular coordinate, see Fig. 3
B_U = fitting parameter, relaxation time for scattering on phonons, K	N_X = phonon transport parameter, defined in Eq. (18), m^{-1}	η = grain-boundary scattering strength, defined by Eq. (3)
C_V = phonon specific heat per unit volume, $J m^{-3} K^{-1}$	$n_{GB,j}$ = number of defects of type j per unit grain-boundary area, m^{-2}	Λ = phonon mean free path = $v_s \tau$, m
D_{RC} = diameter of extended defects, m	p = probability for transmission through grain boundary without scattering	Λ_{AV} = average phonon mean free path, defined in Eq. (29), m
d_D = maximum separation of defective region from grain boundaries, m	q_s = heat flux along layer, $W m^{-2}$	Λ_{GB} = phonon mean free path, grain boundaries, m
d_G = characteristic grain dimension, m	q_{BULK} = heat flux in a homogeneous bulk medium, $W m^{-2}$	Λ_z = linear slope of phonon mean free path with increasing z
$d_{G,n}$ = distance between consecutive grains along phonon trajectory line, m	$S_{A1}, S_{A2}, S_{B1}, S_{B2}$ = parameters for integrand, Eqs. (24)–(26)	Λ_0 = minimum phonon mean free path, m
d_L = layer thickness, m	T = temperature, K	Λ_1, Λ_2 = phonon mean free paths for step nonhomogeneity, m
F_A, F_B = parameters in integrand, m	v_s = average phonon velocity, $m s^{-1}$	$\mu = \cos(\alpha_p)$
h_p = Planck's constant divided by $2\pi = 1.05 \times 10^{-34} J s$	x, y = coordinates along layer, m	σ_j = phonon-scattering cross section of defects of type j , m^2
I_{GB} = distribution function for relating d_G and B_U , m^{-1}	z = coordinate normal to layer, m	τ = phonon relaxation time = Λ/v_s , s
J = total number of types of defects	z_1 = coordinate of step nonhomogeneity, m	τ_{GB} = phonon relaxation time, defects concentrated at grain boundaries, s
j = integer denoting defect type	z_E = acoustic depth, defined in Eq. (17)	τ_D = phonon relaxation time, defects, s
k_B = Boltzmann constant = $1.38 \times 10^{-23} J K^{-1}$	α = angular coordinate, see Fig. 3	τ_U = phonon relaxation time, Umklapp processes, s
m, n = integers used in summation		ω = phonon angular frequency, $rad s^{-1}$

tures above a few degrees Kelvin, the rate of phonon scattering on distinct grain boundaries, which is independent of the phonon frequency, is much smaller than the rate of scattering on other types of defects, e.g., impurities, which depends strongly on the phonon frequency.

An explanation is suggested by the electron micrographs of Heatherington et al. (1990), which show a high concentration of defects within about 2000 Å of grain boundaries. Scattering on defects concentrated at grain boundaries yields anisotropy in layers with columnar grains and has the phonon frequency dependence needed to explain low-temperature conductivity data. Goodson et al. (1994b) used this assumption to relate the effective thermal conductivity along layers to d_G , yielding predictions in reasonable agreement with data for which the local value of d_G is known. This work assumes that in layers of thickness less than a few micrometers, the small grain dimension causes phonon scattering on defects near grain boundaries to dominate over scattering on defects within grains. This allows $1/\tau_D$ in Eq. (1) to be replaced by $1/\tau_{GB}$, which is the scattering rate due to grain boundaries. This work also assumes that $1/\tau_D$ can be treated as homogeneous within a given plane z in solving the phonon transport equation, even though the scattering rate must be higher near grain boundaries. This yields accurate predictions of the heat flux as long as (a) the characteristic length scale of the medium in which transport is analyzed is large compared to d_G and (b) the geometry is such that energy must travel through many grain boundaries, both of which are satisfied here.

This analysis uses two different approximations to relate $1/\tau_{GB}$ to d_G . The first, approximation A, distributes the defects contributed by grain boundaries homogeneously within the plane z . The second, approximation B, assumes that the defects contributed by a grain boundary are located within a distinct distance from grain boundaries, d_D , that is finite but much smaller than d_G . For case B, the scattering rate due to grain boundaries is analyzed considering ballistic phonon transport between boundaries, at which phonons have a probability for transmission without scattering. For both cases, the total scattering rate is calculated using Eq. (1) with the expression for the scattering rate due to grain boundaries derived in the following subsections. Approximation A is most appropriate for $d_G \sim d_D$ and approximation B is most appropriate for $d_G \gg d_D$. The electron micrographs of Heatherington et al. (1990) indicate that $d_D \sim 2000$ Å for grains with $d_G \sim 1$ μm. Since grain dimensions in diamond layers vary between a few tens of nanometers to several tens of micrometers depending on the coordinate z , both approximations are of practical importance.

2.1 Approximation A: Lateral-Homogeneous Distribution of Defects Related to Grain Boundaries. Because grain-boundary scattering is considered geometrically in this work, it is helpful to define a mean free path for scattering on grain boundaries $\Lambda_{GB} = \tau_{GB} v_s$, where $v_s = 1.32 \times 10^4$ m s⁻¹ is the average phonon velocity in diamond. Given approximation A, the inverse of the mean free path can be expanded using

$$[\Lambda_{GB}(z, \omega)]^{-1} = B_V(z) \eta(z, \omega) \quad (2)$$

where B_V is the grain-boundary area per unit volume. The scattering rate determined using Eq. (2) can be applied directly in Eq. (1). The dimensionless grain-boundary scattering strength is defined here,

$$\eta(z, \omega) = \sum_{j=1}^J \sigma_j(\omega) n_{GB,j}(z) \quad (3)$$

where σ is the scattering cross section of defects of type j for phonons of frequency ω . The concentration of defects of type j is the product of the number density of those defects per unit grain-boundary area, $n_{GB,j}$, and B_V . Section 2.2 shows that η is

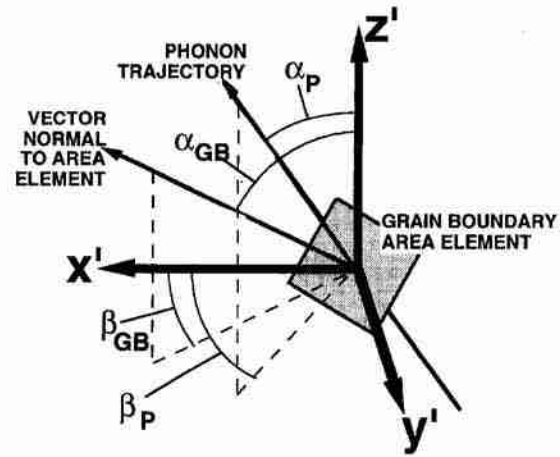


Fig. 3 Angles describing a phonon trajectory and a differential grain-boundary area element

simply related to the probability for phonon transmission through a grain boundary without scattering.

This work derives the relationship between B_V and d_G by assuming that d_G is independent of the direction of the reference line within a given x - y plane. This is not strictly correct for nearly heteroepitaxial layers, which have a large fraction of grain boundaries oriented in the crystallographic directions of the underlying silicon substrates (e.g., Rösler et al., 1993). But the oriented grains prevail only for relatively large values of z , so the present approach is still appropriate. The angles α_{GB} and β_{GB} of the area-element normal vector and a set of relative rectangular coordinates are defined in Fig. 3. The grain-boundary area per unit volume is

$$B_V(z) = 4 \int_0^\pi \int_0^{\pi/2} \frac{I_{GB}(z, \alpha_{GB}, \beta_{GB})}{\sin(\alpha_{GB}) \cos(\beta_{GB})} d\beta_{GB} d\alpha_{GB} \quad (4)$$

where $I_{GB}(\alpha_{GB}, \beta_{GB}) d\alpha_{GB} d\beta_{GB}$ is the number of intersections per unit length of grains of angles α_{GB} and β_{GB} with the x' axis, which serves here as the reference line for the measurement of d_G . The denominator of the integrand in Eq. (4) accounts for the increased contribution of grain-boundary area per unit volume by area elements with normal vectors not parallel to the reference line.

If the grain structure is random, then I_{GB} is

$$I_{GB}(z, \alpha_{GB}, \beta_{GB}) = [\sin(\alpha_{GB})][\sin(\alpha_{GB})|\cos(\beta_{GB})|] \left[\frac{1}{2\pi d_{GB}(z)} \right] \quad (5)$$

The first term in brackets accounts for the fraction of grain-boundary area elements in the x' - y' plane having angle α_{GB} , which decreases to zero with α_{GB} . The second term in brackets accounts for the diminished profiles presented by grain-boundary area elements whose normal vectors are not parallel to the reference line. In layers with columnar grain structure, grain boundaries are predominantly parallel to the z axis but have intersections with x' - y' planes that are randomly oriented. For this case, I_{GB} is

$$I_{GB}(z, \alpha_{GB}, \beta_{GB}) = \left[\delta\left(\alpha_{GB} - \frac{\pi}{2} + \Delta\alpha\right) + \delta\left(\alpha_{GB} - \frac{\pi}{2} - \Delta\alpha\right) \right]$$

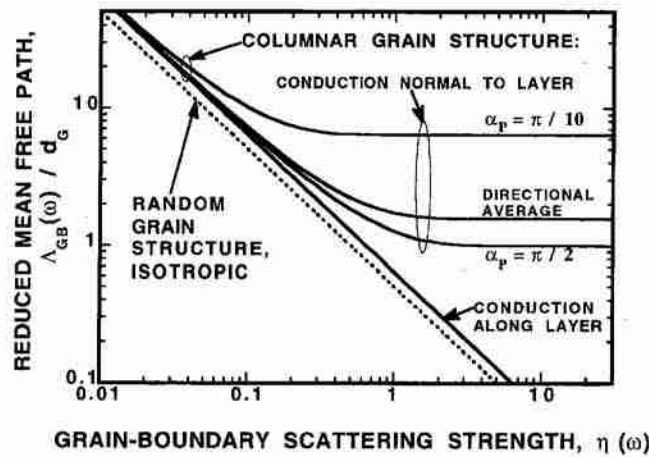


Fig. 4 Phonon mean free path limited by scattering due to grain boundaries

$$\times [|\cos(\beta_{GB})|] \left[\frac{1}{8d_G(z)} \right];$$

$$\sin(\Delta\alpha) = \frac{1}{2} \frac{d}{dz} [d_G(z)] \quad (6)$$

The use of delta functions assumes that grain boundaries have exactly the slope that accommodates the change of d_G with respect to z . Combining Eqs. (2), (4), and (5) yields

$$\frac{\Lambda_{GB}(z, \omega)}{d_G(z)} = \frac{1}{2\eta(z, \omega)} \quad (7)$$

for random grains and

$$\frac{\Lambda_{GB}(z, \omega)}{d_G(z)} = \frac{2}{\pi\eta(z, \omega)} \left[1 + \left(\frac{1}{2} \frac{d}{dz} [d_G(z)] \right)^2 \right]^{-1/2} \quad (8)$$

for columnar grains. The term in brackets in Eq. (8) is usually very near to unity.

The mean free paths for random and columnar grains are plotted in Fig. 4. Practical layers have grain structures between these two extremes. The difference between the two results becomes clear when considering the contribution of each grain boundary to B_V within a cylindrical differential volume element centered about the reference line used to measure d_G . Each intersection of the reference line with the boundary of a columnar grain contributes less to B_V than an intersection with a typical randomly oriented grain boundary.

2.2 Approximation B: Concentration at Grain Boundaries of Defects Related to Grain Boundaries. If d_G is considerably larger than d_D , then approximation A is poorly suited. The error is greatest if the geometry makes it unnecessary for phonons to travel through grain boundaries, which is the case for conduction normal to a layer with a perfect columnar grain structure. Equations (7) and (8) predict that the phonon mean free path, and therefore the thermal conductance for heat transport normal to the layer, can be made arbitrarily small by increasing the grain-boundary scattering strength, η . But if defects related to grain boundaries are restricted to a region near grain boundaries of thickness much less than d_G , then a very large η can at most reduce Λ_{GB} to the order of d_G .

The phonon mean free path to be used for conduction normal to layers with columnar grains is obtained here using approximation B, which assumes that d_D is much smaller than d_G . The probability of phonon transmission through a grain boundary without scattering, p , decreases exponentially with the ratio of the path length of the phonon through the defective region,

which for phonons with normal incidence is $2d_D$, to the phonon mean free path in the defective regions, which is $2d_D/\eta$. This yields

$$p[\eta(z, \omega), \alpha_p - \alpha_{GB}, \beta_p - \beta_{GB}] = \exp \left[\frac{-\eta(z, \omega)}{|\cos(\alpha_p - \alpha_{GB}) \cos(\beta_p - \beta_{GB})|} \right] \quad (9)$$

where the angles α_p and β_p are those of the phonon trajectory shown in Fig. 3. The mean free path of a group of phonons launched with a given trajectory is

$$\Lambda_{GB} = d_{G,0} + \sum_{n=1}^{\infty} d_{G,n} \prod_{m=1}^n p \left[\eta, \alpha_p - \frac{\pi}{2}, \beta_p - \beta_{GB,m} \right] \quad (10)$$

The integer subscripts label the grain boundaries intersected by the phonon trajectory line. The angle $\beta_{GB,m}$ is for the area element of the grain boundary m at the intersection of the trajectory line. The length $d_{G,n}$ is the distance along the trajectory line between grain boundaries n and $n+1$. To obtain an approximate result in closed form, all grain-boundary area elements intersected by the phonon trajectory line are assumed to have the same angle β_{GB} and all $d_{G,n}$ are all assumed to be $d_G/\sin(\alpha_p)$. Equation (10) reduces to

$$\frac{\Lambda_{GB}(z, \alpha_p, \omega)[\sin(\alpha_p)]}{d_G(z)} = \left[1 + \frac{1}{\exp \left[\frac{\pi}{2} \frac{\eta(z, \omega)}{\sin(\alpha_p)} \right] - 1} \right] \quad (11)$$

The coefficient in the exponential function, $\pi/2$, is determined by requiring that approximations A and B yield the same mean free path when $\eta/\sin(\alpha_p)$ is much smaller than unity. In this limit, phonons travel through many grains and the distinction between the two approximations is unimportant. For solutions to the phonon transport equation that assume an isotropic phonon mean free path, it is useful to average Eq. (11) over the solid hemisphere. Numerical integration yields a result within 3 percent of

$$\frac{\Lambda_{GB}(z, \omega)}{d_G(z)} = \frac{\pi}{2} \left[1 + \frac{1}{\exp \left[\left(\frac{\pi}{2} \right)^2 \eta(z, \omega) \right] - 1} \right] \quad (12)$$

Figure 4 shows that Eqs. (11) and (12), which are appropriate for columnar grains, yield longer mean free paths than the results for randomly oriented grains for large η .

2.3 Parameter Values for CVD Diamond. The results derived in Sections 2.1 and 2.2 apply to polycrystalline layers. The results are applied to CVD diamond layers using the phonon-scattering parameters of Graebner (1993) and Goodson et al. (1994b). The first term on the right of Eq. (1) is

$$[\tau_U(x_\omega, T)]^{-1} = A_U(x_\omega)^2 T^3 \exp \left(\frac{-B_U}{T} \right) \quad (13)$$

where $A_U = 640 \text{ s}^{-1} \text{ K}^{-3}$ and $B_U = 470 \text{ K}$. The dimensionless phonon frequency is $x_\omega = h_p \omega / (k_B T)$, where h_p is Planck's constant divided by 2π and k_B is the Boltzmann constant. Imperfections are considered using

$$\sigma_1(x_\omega T) = \frac{V_0^2 x_\omega^4 T^4}{4\pi v_s^4} \left(\frac{k_B}{h_p} \right)^4 \quad (14)$$

for point defects, where $V_0 = 5.68 \times 10^{-30} \text{ m}^3$, and

$$\sigma_2(x_\omega T) = \frac{\pi D_{RC}^6}{4v_s^4} \left(\frac{k_B}{h_p} \right)^4 (x_\omega T)^4 \quad \text{for } x_\omega T < \frac{h_p v_s}{k_B D_{RC}}$$

$$= \frac{\pi (D_{RC})^2}{4} \quad \text{for } x_\omega T \geq \frac{h_p v_s}{k_B D_{RC}} \quad (15)$$

for extended defects, e.g., groups of point defects or small regions with amorphous microstructure. These cross sections vary with phonon angular frequency but not with temperature, so the independent variable is denoted as the product $x_\omega T$. This work uses the values fitted by Goodson et al. (1994b) for the diameter of extended defects, $D_{RC} = 1.4 \times 10^{-9} \text{ m}$, and for the number densities of defects per unit grain boundary area, $n_{GB,1} = 2.2 \times 10^{20} \text{ m}^{-2}$ and $n_{GB,2} = 1.5 \times 10^{18} \text{ m}^{-2}$.

These scattering cross sections do not explicitly account for normal phonon-phonon scattering processes. While normal phonon-phonon scattering does not contribute directly to the thermal resistance, it increases the coupling between phonon scattering at high frequencies and phonons at low frequencies, which effectively increases the resistance. This could be most significant at low temperatures, where normal processes become more important compared to Umklapp processes. The scattering cross sections used here were fitted to thermal conductivity data for thick diamond layers over a large regime of temperatures (Graebner, 1993), and therefore implicitly account for normal processes. A small error may result in the present study of thin layers due to the impact of nonhomogeneity and boundary scattering on the influence of normal processes.

3 Solutions to the Phonon Transport Equation for Nonhomogeneous Layers

The scattering mechanisms discussed in Section 2 govern phonon transport *within* diamond layers. For layers that are sufficiently thin, phonon scattering on the layer boundaries can also be important (e.g., Flik et al., 1992). This necessitates solving the phonon transport equation of Peierls (1955) based on the Boltzmann equation, whose boundary conditions describe the interaction of phonons with the interfaces of a layer. To accommodate the phonon mean free path developed in Section 2, the collision integral in the Peierls-Boltzmann equation must be simplified using the relaxation-time approximation. If the medium is isotropic and the differences between phonon polarizations are neglected, the steady-state form of this equation for the two-dimensional geometry in Fig. 2 reduces to

$$\sin(\alpha_p) \cos(\beta_p) \frac{\partial N(x, z, \omega, \alpha_p, \beta_p)}{\partial x}$$

$$+ \cos(\alpha_p) \frac{\partial N(x, z, \omega, \alpha_p, \beta_p)}{\partial z}$$

$$= \frac{N_0(\omega, T(x, z)) - N(x, z, \omega, \alpha_p, \beta_p)}{\Lambda(z, \omega, T)} \quad (16)$$

where N is the local phonon distribution function and N_0 is the local equilibrium distribution function. An approximate solution approach is to enforce energy conservation within each energy level (e.g., Brewster, 1992), which in this case is distinguished by the phonon angular frequency, ω . This can lead to inaccurate predictions of phonon distribution functions, but it is expected to yield good predictions of the total phonon heat flux. When this method is used, Eq. (16) differs only by a constant of multiplication from the equation of phonon radiative transfer derived by Majumdar (1993). It is useful to define the dimensionless *acoustic depth*

$$z_E(z, \omega, T) = \int_0^z \frac{dz'}{\Lambda(z', \omega, T)} \quad (17)$$

3.1 Transport Along a Layer. Transport in the x direction is due to the following condition:

$$\frac{\partial N_0(\omega, T)}{\partial x} = \frac{\partial N_0(\omega, T)}{\partial T} \frac{\partial T}{\partial x} = N_X \quad (18)$$

where N_X is defined here. For conduction along layers it helps to integrate the Eq. (16) along a line of sight. The position along this line is z_E/μ , where $\mu = \cos(\alpha_p)$. For phonons with $\mu > 0$, i.e., that travel upward in Fig. 2, the integration yields

$$N(z_E, x_0) = N_0[x(z'_E = 0)] \exp\left(-\frac{z_E}{\mu}\right)$$

$$+ \int_0^{z_E} N_0[x(z'_E)] \exp\left(-\frac{z'_E - z_E}{\mu}\right) \frac{dz'_E}{\mu} \quad (19)$$

where

$$N_0[x(z'_E)]$$

$$= N_0(x_0) - N_X[z(z_E) - z(z'_E)] \tan(\alpha_p) \cos(\beta_p) \quad (20)$$

The heat flux is an integral involving the distribution function over all frequencies. The challenge for a nonhomogeneous medium arises due to the nested integral, whose evaluation must carefully consider the variation of the free path in the medium. This work chooses two forms for $\Lambda(z)$ that allow the heat flux to be evaluated analytically:

Linear Nonhomogeneity:

$$\Lambda(z, \omega, T) = \Lambda_0(\omega, T) + z\Lambda_z(\omega, T) \quad (21)$$

Step Nonhomogeneity:

$$\Lambda(z, \omega, T) = \Lambda_1(\omega, T), 0 \leq z < z_1 \quad (22)$$

$$= \Lambda_2(\omega, T), z_1 \leq z < d_L$$

where x_ω is the dimensionless phonon frequency defined in Section 2.3. For the calculations presented in Section 4, a linear dependence is used, as in Eq. (21). This gradual growth is consistent with micrographs of the layers whose transport properties are predicted in the present study. Conduction in some diamond layers may be better modeled using the step change, due to a highly disordered near-interfacial region beneath a region of relatively large grains.

The heat flux for both cases reduces to

$$q_x(x_\omega) = -\frac{dT}{dx} C_V(x_\omega, T) \frac{v_s}{4d_L}$$

$$\times \int_0^{d_L} \int_0^{\pi/2} \sin^2(\alpha_p) \tan(\alpha_p) [F_A + F_B] d\alpha_p dz \quad (23)$$

where d_L is the layer thickness. The specific heat function C_V is from the theory of Debye (Berman, 1976). For linear nonhomogeneity, the parameters F_A and F_B are

$$F_A = z(S_{A1} + S_{A2} - S_{A1}S_{A2}) - \frac{zS_{A1} - \left(\frac{\Lambda_0}{\Lambda_z}\right)S_{A2}}{\mu\Lambda_z + 1} \quad (24)$$

$$F_B = z(S_{B1} + S_{B2} - S_{B1}S_{B2}) - \frac{zS_{B1} - \left(\frac{\Lambda_0}{\Lambda_z} + d_L\right)S_{B2}}{1 - \mu\Lambda_z} \quad (25)$$

where

$$S_{A1} = \left(\frac{\Lambda_0}{z\Lambda_z} + 1 \right); \quad S_{A2} = \left(\frac{z\Lambda_z}{\Lambda_0} + 1 \right)^{-1/\mu\Lambda_z}$$

$$S_{B1} = \left(1 - \frac{\Lambda_0}{z\Lambda_z} - \frac{d}{z} \right); \quad S_{B2} = \left(1 - \frac{z\Lambda_z}{\Lambda_0 + d\Lambda_z} \right)^{1/\mu\Lambda_z} \quad (26)$$

For the step nonhomogeneity,

$$F_A = \mu\Lambda_1 \left[1 - \exp\left(\frac{-z}{\mu\Lambda_1}\right) \right] \quad \text{for } 0 \leq z \leq z_1$$

$$= \mu\Lambda_2 + z \exp\left[\frac{z_1 \left(1 - \frac{\Lambda_2}{\Lambda_1} \right) - z}{\mu\Lambda_2} \right] + \exp\left[\frac{z_1 - z}{\mu\Lambda_2} \right]$$

$$\times \left[(z + \mu\Lambda_1) \left(1 - \exp\left(\frac{-z_1}{\mu\Lambda_1}\right) \right) - z - \mu\Lambda_2 \right]$$

for $z_1 \leq z \leq d_L$ (27)

$$F_B = \mu\Lambda_2 \left[1 - \exp\left(\frac{-z}{\mu\Lambda_2}\right) \right] \quad \text{for } 0 \leq z \leq d_L - z_1$$

$$= \mu\Lambda_1 + z \exp\left[\frac{(d_L - z_1) \left(1 - \frac{\Lambda_1}{\Lambda_2} \right) - z}{\mu\Lambda_1} \right]$$

$$+ \exp\left[\frac{d_L - z_1 - z}{\mu\Lambda_1} \right]$$

$$\times \left[(z + \mu\Lambda_2) \left(1 - \exp\left(\frac{z_1 - d_L}{\mu\Lambda_2}\right) \right) - z - \mu\Lambda_1 \right]$$

for $d_L - z_1 < z \leq d_L$ (28)

It is useful to compare the heat flux calculated here to that along a very thick homogeneous layer, which has a mean free path that is equal to the average of that in the thin diamond layer,

$$q_{\text{BULK}}(x_\omega) = -\frac{dT}{dx} \frac{1}{3} C_V(x_\omega, T) \frac{v_x}{d_L} \int_0^{d_L} \Lambda(z, x_\omega) dz$$

$$= -\frac{dT}{dx} \frac{1}{3} C_V(x_\omega, T) v_s \Lambda_{\text{AV}}(x_\omega) \quad (29)$$

Figure 5 contrasts the ratio of Eqs. (23) and (29) to the solution to the transport equation for a homogeneous medium, which was provided by Fuchs (1938) and Sondheimer (1952). Although these authors actually solved the equation to determine the impact of electron-boundary scattering on the electrical conductivity, the mathematics is completely analogous. When the linear form applies, the average heat flux along the layer can be estimated within 15 percent using the solution of Fuchs (1938) and Sondheimer (1952) and Λ_{AV} . Nonhomogeneity reduces the heat flux along a layer. Conduction in regions with a relatively long mean free path is more strongly impeded by the boundaries than in a homogeneous layer with the same average mean free path, which prevents the heat flux in these regions from completely balancing the low heat flux in regions with a relatively short mean free path. This is most clearly evidenced

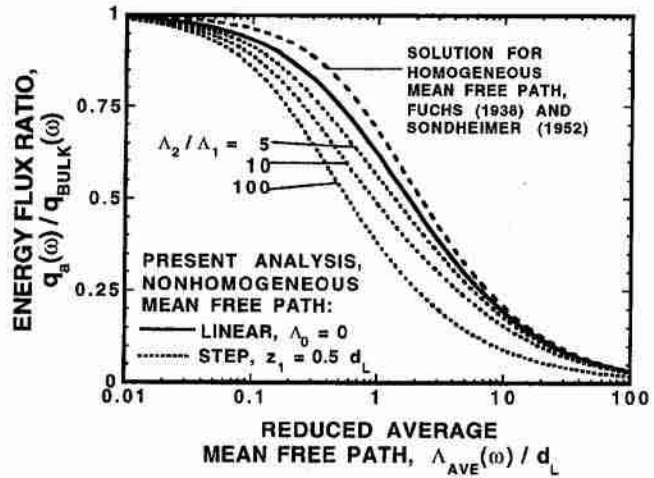


Fig. 5 Reduction of heat flux along a thin layer due to boundary scattering considering a nonhomogeneous mean free path. The bulk heat flux is that for an infinite medium with mean free path equal to the spatially averaged value in the layer.

by the step model, which yields predictions that differ substantially from the homogeneous case.

The effective thermal conductivity along the layer is

$$k_{a,\text{eff}} = -\left(\frac{dT}{dx}\right)^{-1} \int_0^{\theta/T} q_a(x_\omega) dx_\omega \quad (30)$$

where $\theta = 2032$ K is the Debye temperature of diamond.

3.2 Transport Normal to a Layer. The solution to Eq. (16) for conduction normal to a layer is discussed in textbooks for photon transport normal to a gray medium with gray, diffuse boundaries (e.g., Brewster, 1992). The exact solution for the heat flux cannot be written explicitly. But the solution obtained using the diffusion approximation and a slip boundary condition is simple and agrees within a few percent of the exact result. Chen and Tien (1993) used the approximate result to derive the effective thermal conductivity for conduction normal to homogeneous layers. This expression yields predictions that agree with thermal-resistance data for conduction normal to thin amorphous silicon-dioxide layers at low temperatures (Goodson et al., 1994a). For a nonhomogeneous medium,

$$\frac{1}{R_T} = \frac{k_{n,\text{eff}}}{d_L} = \frac{v_s}{3} \int_0^{\theta/T} \frac{C_V(x_\omega, T) dx_\omega}{d_E(x_\omega, T) + \frac{4}{3} \left(\frac{1}{\alpha_0} + \frac{1}{\alpha_1} - 1 \right)} \quad (31)$$

where the dimensionless effective layer thickness d_E is calculated using $z = d_L$ in Eq. (17). The transmission coefficients for phonons into the bounding media, α_0 and α_1 , are calculated using the diffuse mismatch theory of Swartz and Pohl (1989).

This study neglects lateral variation of the phonon scattering rate, which yields a thermal resistance that is homogeneous in the x - y plane. In practical layers, the thermal resistance may vary significantly in the x - y plane due to the higher scattering rate near grain boundaries. Since the resistance is dominated by the regions with smallest grains, nonuniformity would be observable using experimental techniques with resolution finer than d_{G0} . Section 4 compares the thermal resistance predictions with data from a laser-reflectance technique with resolution limited by diffraction to roughly $1 \mu\text{m}$. Since d_{G0} is of the order of 20–50 nm for the layers interrogated here, the comparison is appropriate.

4 Results and Discussion

Figure 6 plots the effective conductivity along layers as a function of the layer thickness at temperatures of 300 and 77 K. The calculations are performed using two different linear slopes relating the grain dimension to z . The data in the figure are for layers in which the reported grain dimension at a given thickness is approximately equal to that used in the calculations. Graebner et al. (1992b) measured the conductivity along several thin layers of varying thickness grown under different conditions. The two layers studied here were grown at nearly the same rate and had almost the same ratios of $d_G(d_L)/d_L$. In order to plot the data in Fig. 6, these two layers are both assumed to have the same linear dependence of d_G on z . The intercept grain dimension, $0.2 \mu\text{m}$, is chosen for consistency with the other grain-dimension function, which was provided by Plamann et al. (1994) for their layers. While both sets of layers were fabricated using a similar microwave-plasma assisted technique, the process of Plamann et al. (1994) yielded a microstructure with finer grains. The agreement between the predictions and data at room temperature is achieved using no fitting parameters. The grain-boundary scattering strength η used here was determined during a prior research project by fitting the conductivity of diamond layers thicker than $50 \mu\text{m}$ (Goodson et al., 1994b). This lends support to the assumption that η is independent of z .

Figure 6 shows the importance of obtaining a rapid increase of grain dimension with the layer thickness. The conductivity of layers with larger grains increases far more rapidly with increasing z , which can allow a total required conductance along a layer to be achieved using less diamond. The conductivities predicted in Fig. 6 are much less than that of high-quality thick CVD diamond layers at both 77 K and 300 K, which exceeds $2000 \text{ W m}^{-1} \text{ K}^{-1}$. The reduction is most severe at cryogenic temperatures. At 77 K the phonon mean free path is limited by scattering on grain and layer boundaries and the phonon specific heat is much less than that at room temperature. This causes the conductivities at 77 K of the diamond layers to be more than an order of magnitude less than those at room temperature, although the bulk conductivity has the opposite behavior.

For conduction normal to diamond layers, the grain size at the interface is very important. Figure 7 predicts the thermal resistance using a relationship between d_G and z that is based on top-view electron micrographs of layers experimentally investigated by Goodson et al. (1995b), one of which is shown in Fig. 1. The data agree well with the predictions if a random grain structure is assumed and the diamond grain dimension

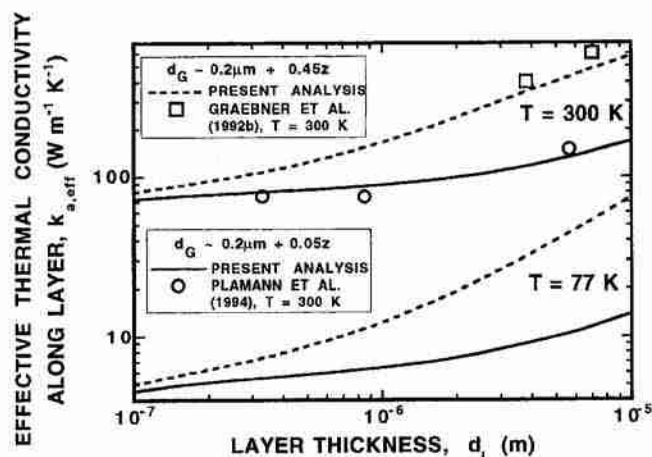


Fig. 6 Effective conductivity along thin nonhomogeneous diamond layers considering two relationships between the grain size and thickness. The data are for layers whose reported grain dimensions for a given thickness are the same as those used in the respective calculations.

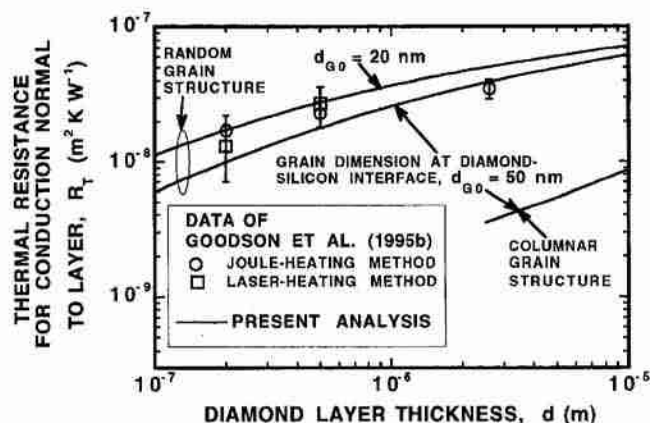


Fig. 7 Thermal resistance for conduction normal to thin metallized diamond layers on silicon

near the interface is assumed to be between 20 and 50 nm. These assumptions are consistent with top-view micrograph of the layers in the study, although the grain structure of the thickest layer appeared to be at least partially columnar. A columnar grain structure improves conduction in the direction normal to the layer compared to that for a random grain structure, as shown in the predictions in Fig. 7. This may explain the overprediction of the data for the thickest layer by the analysis that assumes randomly oriented grains.

The scattering theory in Section 2 can be applied independently of the solution to the transport equation to estimate the local conductivity in layers of thickness greater than $1 \mu\text{m}$ at room temperature as a function of the grain dimension d_G . Using approximation A with Eqs. (29) and (30), the diamond conductivity at 300 K is fit within 6 percent by

$$k(d_G) = (2169 \text{ W m}^{-1} \text{ K}^{-1}) \left[1 - \frac{6.785 \times 10^{-6} \text{ m}}{d_G + 6.785 \times 10^{-6} \text{ m}} \right] \quad (31)$$

5 Conclusions

This research uses phonon transport theory to relate the conductivity of thin diamond layers to the size and orientation of their grains. The agreement between predictions and data lend support to the hypothesis that defects concentrated near grain boundaries limit the mean free path of phonons in thin diamond layers. Future deposition research must target the most rapid increase in grain dimension with thickness during layer growth. The different mean-free-path expressions derived here for conduction along and normal to layers with columnar grains should be used to model the anisotropy measured in thick diamond layers (Graebner, 1993).

This work shows that contemporary layers have the potential to provide electrical insulation without the problematic thermal resistance resulting from conventional dielectrics, e.g., amorphous silicon dioxide. A major task required to realize this potential is to minimize the thermal boundary resistances between diamond and neighboring materials, which can result due to disordered interfacial layers or to incomplete contact.

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