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Predictions of thermal boundary conductance for systems of disordered solids and interfaces

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As microsystems grow in their complexity, the number of material layers increases even as the thickness of these layers decreases. As a consequence, energetic transport through material intersections, the so-called thermal boundary conductance (TBC), becomes a greater contributor to the total thermal response of the system as a whole. Consequently, methods are sought that allow for insight into the mechanisms determining the efficiency of this transport, while simultaneously providing predictions with minimal computational investiture. In response, the current study extends the often employed diffuse mismatch model (DMM) to account for disorder that is frequently present in the materials making up the interface as well as the boundary itself. By applying assumptions regarding the scattering rates and mean free paths of phonons within a disordered solid, the resulting modifications of the spectral density of states induce changes in both the number and ratio of forward scattered phonons incident on a surface, and hence predictions of the TBC. Combining these assumptions with an accounting of the distance over which disorder persists, the newly implemented disorder DMM (δ-DMM) is shown to be more capable of predicting the TBC over a range of temperatures and material systems. Additionally, the model demonstrates that TBC is dependent on not only on the material properties but also on the morphology of these materials and the nature of their union. © *2009 American Institute of Physics*. [doi:10.1063/1.3267496]

I. INTRODUCTION

Energetic transport across material boundaries is increasingly relevant as devices grow in their complexity, multiplying not only the number of interfaces but also the interfacial influence on the system performance as a whole. Devices designed for such varying applications as thermoelectrics,[1] high power electronics,[2] and thermal interface materials[3] are significantly affected by the efficiency in which thermal energy crosses the numerous interfaces that are embedded within these structures. Consequently, analytical tools that highlight the central physics determining the rate by which thermal energy crosses material interfaces, i.e., the thermal boundary conductance (TBC), are useful in their ability to not only quickly estimate performance but to also elucidate the mechanisms and processes by which device designers may leverage this transport.

Widely utilized, one such analytical tool is the diffuse mismatch model (DMM) originally developed by Swartz and Pohl.[4] The model capitalizes on the fact that at room temperature and above, the probability of diffuse phonon scattering is exceedingly large.[5] Thus, by assuming that all vibrational modes are indeed diffusively scattered, predictions of the TBC are made possible by counting both the flux of phonons reaching the boundary as well as the percentage of these phonons that are transmitted across the interface. Applying this approach, the DMM is more straightforward in its implementation and interpretation than more rigorous counterparts such as lattice dynamical and molecular dynamics simulations, making it a prime tool in not only the analysis of experimental results[6–8] but also the prediction of an entire system's thermal behavior.[9–11]

Despite its pronounced implementation, experimental results oftentimes vary in comparison with predictions obtained with the DMM by over an order of magnitude.[12] A host of factors, all of which are not accounted for in the DMM, has been attributed to these large differences including phonon mean free path,[13] inelastic phonon scattering,[14] and the quality of both the constituent materials[6,15] as well as the resulting interface.[16] In response, several investigations extended the DMM in order to more accurately account for each one of these factors by including a more realistic phonon dispersion,[17,18] inelastic transport mechanisms,[19] and multiple phonon scattering events.[20]

While each of these extensions enhances the capability of TBC predictions, their implementation, like the DMM, rests on the supposition that the materials involved in the transport are crystalline. The nature of a real, imperfect, interface is one of disorder, however, in which the assumed planar intersection of each material is, in reality, a finite volume of intermixing. This region of intermixing, defined here as an interphase, can disrupt both the composition of the lattice as well as its spatial arrangement for nanometers at a time, causing an alteration in the nature of the energy transport.[16] In addition, the metal films at the center of many interfacial transport studies themselves contain a substantial degree of disorder. This disorder arises due to the nature of thin film deposition, typically realized through a sputtering or evaporation technique, in which the resulting metal is nanocrystalline or may even be completely amorphous.[21–24] The disruption in the crystallinity of the film, in turn, induces

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changes in the material's vibrational characteristics that modify the nature of the thermal transport. In total, these distinctly noncrystalline characteristics of both the film and the interfacial region highlight the need to consider the ramifications of disorder on the thermal boundary conductance.

To predict the thermal transport within disordered solids, Cahill and Pohl (CP model)[25] assumed that each phonon scatters at a rate of $\frac{1}{2}$ the period of its oscillation, as was originally proposed by Einstein.[26] By then incorporating this assumption into a Debye framework, the CP model is able to accurately predict the thermal conductivity for a variety of amorphous materials. However, as this disordered approach only addresses the rate of scattering and not the nature of the scattered species—i.e., the phonons—the methodology provides no change in the prediction of TBC as it, like the DMM, assumes that the material responds in a Debye fashion. Recently, an alternative approach has been proposed by which all modes are assumed to, as in the CP model, scatter at a rate of $\frac{1}{2}$ the period of its oscillation but, unlike the CP model, have these scattering events separated by only the interatomic distance due to disorder.[27,28] This second constraint removes the equality between the disordered material's phase and group velocities, resulting in a change in the density of states (DOS) within the frequency domain. The alteration in the spectral DOS, in turn, leads to more accurate predictions of both the thermal conductivity as well as the heat capacity. Moreover, the change has distinct consequences for the interfacial transport considered here.

As stated before and further discussed in Sec. II, the DMM predicts the TBC by counting the amount and type of phonons reaching an interface, while simultaneously determining which of these phonons is capable of forward scattering across the boundary. To determine those phonons that are transmitted, the DMM compares how well the DOS between the materials on either side of the interface "spectrally match." Hence, an alteration in the spectral DOS will necessarily change the number of forward scattered phonons and the resulting prediction of TBC, thus providing a manner to evaluate the effect of disorder on the interfacial transport.

With this motivation, the current study examines the effects of disorder on the TBC from contributions both within a film and the finite region of intermixing that comprises a real imperfect interface. The analysis takes place through a modification of the DMM that arises in conjunction with alterations in the DOS evolving from the assumption of a highly disordered film and region of disorder.[27,28] To account for the finite interphase, a two-interface approach like that adopted by Beechem *et al.* is employed where three thermal conductivities in series—(i) TBC of film/interphase, (ii) interphase conductivity, (iii) TBC of interphase/substrate (see Fig. 1)—are utilized to obtain the conductance of the boundary system as a whole.[20] The treatment itself begins with a derivation of the traditional DMM, followed by an explanation of the modifications that become necessary when the materials and interface are assumed to be highly disordered. Subsequently, the efficacy of the newly described disordered diffuse mismatch model (δ-DMM) is then examined by applying it to predict the TBC on a series of the metal film/dielectric interfacial systems that have been previously examined experimentally.

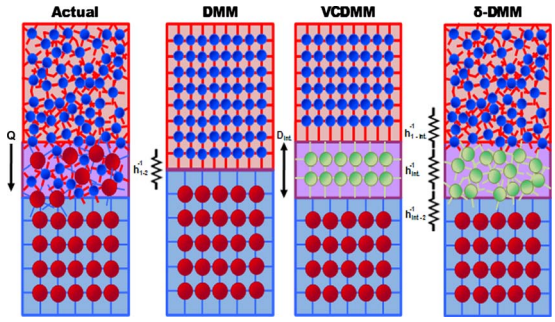

FIG. 1. (Color online) Schematic representation of an "actual" interface subjected to a flux of heat, Q, composed of a disordered film on a crystalline substrate having a region of disorder with thickness, D_{Int}. The DMM predicts the TBC of the system by assuming that both material layers are crystalline and that the interface between them is perfect. To account for both the disorder of the system and the finite thickness of the interphase, the δ-DMM evaluates the conductance in three separate regions: (a) film/interphase interface-$h_{1\text{-Int}}$ (b) interphase-h_{Int} and (c) interphase/substrate boundary-$h_{\text{Int-2}}$. Unlike previous extensions of the DMM such as the virtual crystal approach (VCDMM), the current methodology does not treat the system as entirely crystalline, which allows for both the reduction in free parameters and a greater capability to predict the temperature dependent response of the system.

II. DERIVATION OF THE DMM

Predictions of the thermal boundary conductance begin by quantifying the flux of energy incident on the boundary. It is typically assumed that lattice vibrations (phonons) are the sole carriers of energy across the interface for any system other than that of a metal placed adjacent to another metal.[4,29] Thus, as there is little coherence between the lattice modes when the material layers are thick and the interface is rough,[30] the vibrational modes may be viewed as particles allowing for the flux of energy reaching the interface from side 1, Q_1, to be quantified using the equation of phonon radiative transfer,

$$Q_1 = \sum_j \int_{\Omega=2\pi} \int_0^{\omega_{c1,j}} \mu I_{1,j}(z,t,\omega,v,\Omega) d\omega d\Omega, \tag{1}$$

where μ is the cosine between the wavevector of the mode and the direction perpendicular to the interface, I is the directional spectral intensity of phonons per unit area which is necessarily a function of the distance from the interface, z, time, t, phonon frequency and group velocity, ω and v, and solid angle, Ω. To account for the entirety of the energy impinging on the surface, all participating phonon modes must be quantified. This is accomplished by integrating each phonon branch over the total solid angle of the hemisphere defining material 1 up to its maximum cut-off frequency, $\omega_{c1,j}$, and then summing these contributions over all polarizations, j.

For a finite TBC, the amount of energy that is transmitted through the boundary will be less than the total amount reaching the interface. The ratio of this transmitted energy is represented using the transmission coefficient, α_{12}, where the

subscripts indicate that the energy moves from material 1 to material 2. Using this ratio, the thermal boundary conductance, h_{TBC}, may then be defined as

$$h_{\mathrm{TBC}} = \frac{\partial Q_1}{\partial T} = \frac{\partial}{\partial T}\left[\sum_j \int_{\Omega=2\pi}\int_0^{\omega_{c1,j}} \mu I_{1,j}(z,t,\omega,v,\Omega) \cdot \alpha_{12}(z,t,\omega,v,\Omega)d\omega d\Omega\right]. \quad (2)$$

In the limit of diffuse transport under steady state conditions, all terms lose their directional dependence, allowing for the expression to be integrated over the hemispherical solid angle making up material 1,

$$h_{\mathrm{TBC}} = \frac{\partial}{\partial T}\left[\sum_j \pi \int_0^{\omega_{c1,j}} \mu I_{1,j}(z,t,\omega,v) \cdot \alpha_{12}(z,t,\omega,v)d\omega\right]. \quad (3)$$

Additionally, the diffuse assumption also allows for the interface to be considered a completely thermalizing, or black, boundary.[31] As such, the intensity of phonons may be described as

$$I_{1,j}(\omega,v,T) = \frac{1}{4\pi}\hbar\omega v_j N_j(\omega,T), \quad (4)$$

where $\hbar$ is the modified Planck's constant, T is the temperature, and N_j is the number of participating phonon modes defined using the product of the phonon DOS, $g_{1,j}(\omega)$, and the Bose–Einstein distribution function, $f(\omega,T)$. By then assuming that the solid exhibits a Debye response, the DOS is given by, $g_{1,j}(\omega)=\omega^2/2\pi^2 v_{1,j}^3$, allowing for Eq. (3) to be evaluated as

$$h_{\mathrm{TBC}} = \frac{1}{8\pi^2}\sum_j v_{1,j}^{-2}\int_0^{\omega_{c1,j}} \frac{\hbar^2\omega^4}{k_B T^2} \times \frac{\exp\left(\frac{\hbar\omega}{k_B T}\right)}{\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^2}\alpha_{12}(\omega,v,T)d\omega, \quad (5)$$

where k_B is the Boltzmann constant and the cut-off frequency is defined from Debye theory as $\omega_{1c,j}=v_{1,j}(6\pi^2 n_1/\beta)^{1/3}$, where n_1 is the atomic density and β is the number of atoms per primitive cell. By including β, only the acoustic phonon modes are assumed to contribute to the transport, an assumption that becomes increasingly critical as atoms grow more complex in their basis.[1]

As the scattering is, again, assumed to take place in a completely diffusive manner, upon interacting with the interface the phonons will lose all information as to the direction, phase, polarization, energy, and even the material from which they originated. Furthermore, since the interface acts as a thermalizing black body, the interfacial region may also be assumed to be in a state of local equilibrium. Thus, by assuming all scattering events occur elastically, a fact that is valid only in the limit of materials having similar vibrational characteristics,[32] then the flux of phonons of a given energy leaving one side of the interface must be equal to the flux leaving the other side as well. This is a statement of detailed balance and allows for the evaluation of the transmission coefficient.[4]

The flux of phonons of a given energy, ω, leaving side 1 may be quantified by summing over all polarizations and multiplying the number of participating phonons by the speed at which they propagate. Mathematically, this flux, $P^{12}(\omega)$, is written through an evaluation of the DOS and the distribution function as

$$P^{12}(\omega) = \sum_j v_{1,j} g_{1,j}(\omega) f(\omega,T)\alpha_{12}(\omega,v,T) = \frac{1}{2\pi^2}\sum_j \frac{\omega^2}{v_{1,j}^2}\frac{1}{\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^2}\alpha_{12}(\omega,v,T). \quad (6)$$

Additionally, an equivalent flux, $P^{21}(\omega)$, may be written for the flux of phonons leaving side 2 using the same expression as Eq. (6) with appropriate modifications made for the velocities and the transmission coefficient for phonons scattering from side 2 to 1, $\alpha_{21}(\omega,v,T)$. Recognizing that since phonons scattering with the interface lose all relation to the side from which they originated, the likelihood of reflection on one side must be equivalent to the probability of scattering from the other side, i.e., $\alpha_{21}(\omega,v,T)=r_{12}(\omega,v,T)$. This fact allows for the transmission ratios on either side to be linked via the relation $\alpha_{12}(\omega,v,T)=1-\alpha_{21}(\omega,v,T)$ since $\alpha_{12}(\omega,v,T)+r_{12}(\omega,v,T)=1$. Thus, the phonon fluxes may be equated using Eq. (6) to produce a relation that is dependent on only a single transmission coefficient,

$$\frac{1}{2\pi^2}\sum_j \frac{\omega^2}{v_{1,j}^2}\frac{1}{\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^2}\alpha_{12}(\omega,v,T) = \frac{1}{2\pi^2}\sum_j \frac{\omega^2}{v_{2,j}^2}\frac{1}{\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^2}[1-\alpha_{12}(\omega,v,T)]. \quad (7)$$

Equation (7) may be subsequently solved for $\alpha_{1-2}(\omega,v,T)$ to produce the traditional transmission coefficient of the DMM, and hence the capability to evaluate the TBC,

$$\alpha_{12,\mathrm{DMM}} = \frac{\sum_j v_{2,j}^2}{\sum_j v_{2,j}^2 + \sum_j v_{1,j}^2}. \quad (8)$$

It is of note that the assumptions regarding diffuse elastic scattering and a Debye solid lead to a transmission ratio in the DMM that is independent of both temperature and frequency.

III. PREDICTIONS OF TBC FOR DISORDERED FILMS AND INTERFACES

For many microelectronic applications, a particularly relevant interface is composed of a metal film that is fabricated upon a thicker dielectric substrate. While the substrate itself is most often highly crystalline, the metal film may contain a significant level of disorder due to the evaporative

or sputtering procedures oftentimes employed in their application.[16] In addition, nearly all interfaces, regardless of the film fabrication, exhibit a high degree of disorder surrounding the boundaries between the materials. This interfacial disorder arises due to interdiffusion and defects associated with the material processing, leading to a distinctly noncrystalline layer that may be of the order of tens of nanometers in thickness. Hence, this disordered interfacial region, termed for simplicity as the interphase, may have attributes that are far different than any of the constituent materials and thus may itself be thought of as an additional material layer. Therefore, the total TBC may be characterized as having contributions from three distinct regions (see Fig. 1): (a) interfacial conductance between the film (material 1) and the interphase ($h_{1\text{-int}}$), (b) conduction through the finite thickness of the interphase (h_{int}), and (c) interfacial conductance through the interphase and into the substrate (material 2) ($h_{\text{int-2}}$).[20]

Using such a formulation, heat is assumed to flow from the metal film, into the interphase, and then through the dielectric substrate in a fashion that is analogous to the majority of TBC measurements including those considered as part of this study (see Fig. 1). The direction of heat flow necessitates consideration as the model developed here, like the DMM, predicts the TBC in a fashion that is dependent on the orientation of the transport.[33] To then obtain an estimation for the total TBC of a disordered interfacial system, subsequent subsections will be focused on obtaining predictions for each of these different conductances that determine the efficiency of the transport as it moves from the metal film and into the dielectric substrate.

A. Thermal boundary conductance between two disordered solids

As both the metal film and the interphase region are considered in the limit of heavy disorder,[35] the DMM must be modified to account for the changes in the vibrational characteristic of the solid that naturally occur with a loss in crystallinity. These changes were recently addressed by the authors through an extension of Cahill and Pohl's model[25] utilized in the prediction of thermal conductivity within disordered solids.[27,28] The extension assumes that each phonon mode is scattered at a rate of $\frac{1}{2}$ its period of oscillation with these scattering events separated by the interatomic spacing of the material due to the high level of disorder. Most relevant from a TBC perspective, these assumptions require an inequality between the phase and group velocities of the phonon modes. This fact, in turn, significantly alters the DOS from that of a Debye solid, as is shown below,

$$g_{D,j} = \frac{n_D^{2/3}}{2v_{D,j}}, \tag{9}$$

where $g_{D,j}$ is the polarization specific density of DOS for a disordered solid and $v_{D,j}$ is the group velocity that is assumed to be the speed of sound within the material.

This change in the DOS has significant implications in the determination of TBC as it will modify the phonon intensity and, hence, Eq. (5), as shown below,

$$h_{DD} = \frac{n_1^{2/3}}{8}\sum_j \int_0^{\omega_{c1,j}} \frac{\hbar^2\omega^2}{k_B T^2} \times \frac{\exp\left(\frac{\hbar\omega}{k_B T}\right)}{\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^2}\alpha_{12}(\omega,v,T)d\omega, \tag{10}$$

where it is assumed that material 1, the metal film, is a disordered solid. In addition, the transmission ratio will be modified by the change in the nature of the solid as well. To calculate the modified transmission ratio, recognize that even in the consideration of disordered solids, diffuse scattering requires that the flux of phonons leaving either side of the interface remain equivalent. Therefore, Eq. (7) may be leveraged to find the transmission ratio once again through the use of, now, the modified DOS. If both materials are assumed to be disordered as will be the case between the film and the interphase, this modified DOS must be incorporated on either side of the relation, leading to

$$\sum_j v_{1g,j}\frac{n_1^{2/3}}{2v_{1g,j}}\frac{1}{\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^2}\alpha_{1\text{-int}}(\omega,v,T) = \sum_j v_{\text{int}g,j}\frac{n_{\text{int}}^{2/3}}{2v_{\text{int}g,j}}\frac{1}{\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^2} \times[1-\alpha_{1\text{-int}}(\omega,v,T)], \tag{11}$$

where $\alpha_{1\text{-int}}(\omega,v,T)$ is the transmission ratio between the disordered film and the disordered interphase (corresponding to the subscript "int"), or more generally, any two disordered solids. This transmission ratio may then be obtained using Eq. (11),

$$\alpha_{1\text{-int}} = \frac{n_{\text{int}}^{2/3}}{n_{\text{int}}^{2/3}+n_1^{2/3}}, \tag{12}$$

where n_{int} is the atomic density of the interphase region determined in Sec. III B. With the transmission ratio now obtained, the TBC may be expressed for the boundary between two disordered materials via an expansion of Eq. (10),

$$h_{DD} = \frac{n_1^{2/3}n_{\text{int}}^{2/3}}{8(n_{\text{int}}^{2/3}+n_1^{2/3})}\sum_j \int_0^{\omega_{c1,j}} \frac{\hbar^2\omega^2}{k_B T^2}\frac{\exp\left(\frac{\hbar\omega}{k_B T}\right)}{\left[\exp\left(\frac{\hbar\omega}{k_B T}\right)-1\right]^2}d\omega. \tag{13}$$

In the case of a metal/dielectric system, there is an additional resistance to thermal transport that evolves as a consequence of the primary energy carriers being electrons within the metal and phonons in the dielectric. Majumdar and Reddy[36] accounted for this resistance through development of the relation $h_{\text{e-ph}}=(\sqrt{Gk_m})^{-1}$, where $h_{\text{e-ph}}$ is the electron-phonon coupling conductance, G is the experimentally determined electron-phonon coupling factor,[37] and k_m is

the *lattice* thermal conductivity of the metal. To determine the lattice thermal conductivity of the disordered metal, the expression first proposed by Hopkins and Piekos[28] to predict the thermal conductivity of disordered films is employed and reproduced below,

$$k_m = \left(\frac{\pi^2}{36\beta}\right)^{1/3} n_1^{2/3} k_B \sum_j \frac{T}{\Theta_j} v_{1g,j} \int_0^{\Theta_j/T} \frac{x^2 \exp(x)}{[\exp(x)-1]^2} dx, \tag{14}$$

where $x=\hbar\omega/k_BT$ and the polarization dependent Debye temperature is defined as $\Theta_{1,j}=\hbar\omega_{1c,j}/k_B$. With all the needed parameters now obtained, the total TBC of the metal/interphase boundary may be evaluated through summing of the individual conductivities,

$$h_{1\text{-int}}^{-1} = h_{DD}^{-1} + h_{\text{e-ph}}^{-1}. \tag{15}$$

B. Conductivity through the interphase

After heat is transported across this initial interface, the imperfect nature of the boundary requires that energy propagate through the finite thickness of the disordered interphase. As a consequence, it is necessary to determine the thermal conductivity of this interphase in order to calculate the TBC of the system as a whole. This may be accomplished through implementation of Eq. (14) as this region is assumed to be extremely disordered in accordance with the original development of the relation. To evaluate Eq. (14), material properties of the interphase are obtained through averaging of the individual values from the solids on either side of the interface. Rather than attempting to simulate a given percentage composition as has been done previously,[20] averaging the properties both simplifies the model through the removal of a free parameter and also guarantees the maximum level of disorder in accordance with the conductivity model itself. Using this procedure, the conductance may be quantified by consideration of the thickness over which the disorder persists, D_{int},

$$h_{\text{int}} = \frac{k_{\text{int}}}{D_{\text{int}}}. \tag{16}$$

C. TBC between a disordered and a crystalline solid

Upon propagating through the interphase region, the final component determining the system's TBC is encountered at the boundary between the interphase and the crystalline substrate. To calculate the TBC of this "second interface," Eq. (10) must be modified to account for the fact that the carriers are emerging from the disordered interphase region and attempting to enter a crystalline solid (material 2),

$$h_{\text{dc}} = \frac{n_{\text{int}}^{2/3}}{8} \sum_j \int_0^{\omega_{\text{cint},j}} \frac{\hbar^2\omega^2}{k_BT^2} \times \frac{\exp\left(\frac{\hbar\omega}{k_BT}\right)}{\left[\exp\left(\frac{\hbar\omega}{k_BT}\right)-1\right]^2} \alpha_{\text{int-2}}(\omega,v,T)\, d\omega. \tag{17}$$

To acquire the transmission ratio, the phonon fluxes leaving each material may again be equated. It is of note that the crystalline solid, as in the DMM, is assumed to respond in a Debye fashion and so will have a different DOS than that of the interphase. Explicitly, this is seen via the relation below,

$$\sum_j v_{\text{intg},j} \frac{n_{\text{int}}^{2/3}}{2v_{\text{intg},j}} \frac{1}{\left[\exp\left(\frac{\hbar\omega}{k_BT}\right)-1\right]^2} \alpha_{\text{int-2}}(\omega,v,T) = \frac{1}{2\pi^2} \sum_j \frac{\omega^2}{v_{2,j}^2} \frac{1}{\left[\exp\left(\frac{\hbar\omega}{k_BT}\right)-1\right]^2} [1-\alpha_{\text{int-2}}(\omega,v,T)]. \tag{18}$$

Solving for the transmission ratio yields,

$$\alpha_{\text{int-2}}(\omega) = \frac{\sum_j \frac{\omega^2}{v_{2,j}^2}}{\sum_j \frac{\omega^2}{v_{2,j}^2} + 3\pi^2 n_{\text{int}}^{2/3}}, \tag{19}$$

which allows for Eq. (17) to be expanded into its final form describing the thermal boundary conductance between a disordered and crystalline solid,

$$h_{\text{int-2}} = h_{\text{dc}} = \frac{\hbar^2 n_{\text{int}}^{2/3}}{8k_BT^2} \sum_j \int_0^{\omega_{\text{cint},j}} \omega^4 \frac{\exp\left(\frac{\hbar\omega}{k_BT}\right)}{\left[\exp\left(\frac{\hbar\omega}{k_BT}\right)-1\right]^2} \times \left(\frac{\sum_j \frac{\omega^2}{v_{2,j}^2}}{\sum_j \frac{\omega^2}{v_{2,j}^2} + 3\pi^2 n_{\text{int}}^{2/3}} \right) d\omega. \tag{20}$$

Having now an expression for each of the three components of the conductance, the total TBC of the system may be found through summing the individual contributions allowing for the δ-DMM to take on its final form,

$$\text{TBC}^{-1} = h_{1\text{-int}}^{-1} + h_{\text{int}}^{-1} + h_{\text{int-2}}^{-1}. \tag{21}$$

It is of note that in the case of energy propagating between a disordered and a crystalline solid, the transmission ratio is dependent on the phonon frequency. This is unlike that predicted for either the traditional DMM or the examination of the interface between two disordered solids. Physically, this occurs due to the fact that the DOS within the disordered solid is spectrally constant, whereas that of the

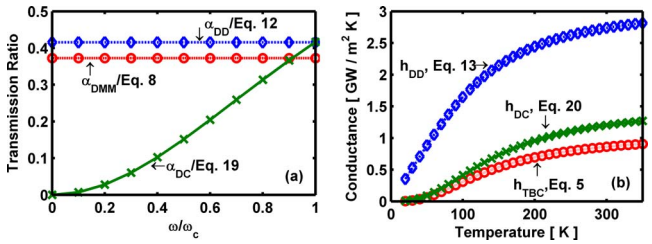

FIG. 2. (Color online) (a) Spectral dependence of the transmission ratio for three types of boundaries: crystal/crystal, disorder/disorder, and disorder/crystal. While the ratio for materials having a similar morphology is spectrally constant, the boundary between a crystalline solid and one exhibiting disorder exhibits a transmission ratio that increases with frequency due to the differences in the spectral DOS between the materials. (b) Variations in the transmission ratio with disorder induce increases in the boundary conductance as the interface becomes inherently less defined. The overall TBC of the entire system [Eq. (21)], however, does not increase with disorder as energy must propagate through the finite thickness of the interphase as well.

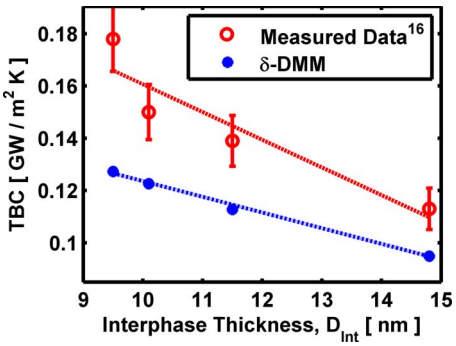

FIG. 3. (Color online) Comparison of thermal boundary conductance measurements made on a series of Cr/Si interfaces [Hopkins *et al.* (Ref. 16)] with predictions obtained from the δ-DMM. Predictions using the δ-DMM correlate far better than those obtained using the standard DMM (DMM of Cr/Si=0.86 GW/m^2 K), while capturing the dependence of interfacial conductance on the thickness of disorder.

crystalline Debye solid increases with frequency. Thus, as more modes become available at higher energies in the crystal there is associated increase within the disordered solid. In order for the flux to then be equal, the ratio of transmitted modes must be modified accordingly.

This fact is illustrated in Fig. 2(a) where the transmission ratio for each of the three types of boundaries examined—crystal/crystal [Eq. (8)], disorder/disorder [Eq. (12)], and disorder/crystal [Eq. (19)]—are compared to one another as a function of phonon frequency for a chromium/silicon (Cr/Si) interface. For the cases where the materials are similar in their morphology, i.e., either both crystalline or both disordered, the transmission ratio is spectrally constant. On the other hand, if only one of the solids exhibits disorder, the resulting transmission ratio increases with phonon energy for the reasons mentioned above. These alterations in the transmission ratio, in turn, necessarily modify the resulting boundary conductance between the two materials, as shown in Fig. 2(b). The disorder results in an increase in the efficiency by which energy is transported from one material to another. The greater efficiency arises in direct relation to the disorder as the solid's nonregularity will necessarily reduce the defined demarcation between materials that serves as the origin of the TBC itself. In effect, the disorder slows the abruptness of the system's change from one material to another, causing an associated reduction in the resistance to energy transport. It should not be inferred, however, that the total TBC of the system will also be enhanced in conjunction with the disorder [see Eq. (21)]. Rather, the same transport enhancing irregularity that disrupts the definition of the boundary also significantly curtails energy transfer as propagation must also occur through this finite region of disorder. Thus, although energy is better able to transition in disordered systems, the total TBC of the system will be seen to actually reduce due to the finite thickness of the interphase.

IV. MODELING TBC IN DISORDERED SYSTEMS USING THE δ-DMM

To evaluate the capability of the newly proposed methodology, a series of previously characterized chromium/silicon (Cr/Si) interfaces was examined using the δ-DMM.[16] The Cr/Si system was chosen due first to the fact that the acoustic mismatch between the materials is small and as such the assumption of transport dominated by elastic scattering retains its validity.[32] More importantly for this analysis, however, is the fact that the previous investigation measured the TBC as a function of the interfacial disorder, thereby allowing for a prescribed, rather than fitted, value of D_{Int} to be utilized in the evaluation of the model. Finally, transmission electron microscopy investigations of the system indicated significant disorder within both the metallic Cr and the disordered interphase region further justifying the incorporation of the current approach in the analysis.

To implement the δ-DMM, material properties for both the Cr and Si were obtained from Ref. 4, while the electron/phonon coupling factor needed in the evaluation of $h_{\mathrm{e\text{-}ph}}$ is gathered from Ref. 37. Shown in Fig. 3 is an evaluation of the δ-DMM [Eq. (21)] for the Cr/Si system where the thickness of the interphase (D_{int}) utilized in the model was determined by the Auger electron microscopy measurements of Hopkins *et al.*[16] By incorporating the measured thickness of the interphase, the δ-DMM contains no free parameters allowing for a direct comparison between theory and experiment. The values obtained using the model correlate well with those acquired experimentally and accurately capture, unlike the DMM, the trend of decreasing TBC with increasing interphase thickness, thus giving confidence to the applicability of the disordered methodology.

In addition to providing an experimental comparison with the model, Fig. 3 also offers insight into the sensitivity of the δ-DMM to the interphase thickness, D_{Int}, as this is the only parameter varied in the analysis of the Cr/Si system. Upon examination, it is found that the TBC is reduced by approximately 25% upon an increase in the interphase thickness from 9.5 to 14.8 nm, leading to roughly a 5% TBC decrease per nanometer of the interfacial disorder. Similar reductions in the TBC between 4%/nm and 5%/nm, are also found for the other material systems considered later in this study, indicating that the relative dependence of the TBC on the interphase thickness is largely independent of the materials involved.

A similar dependence of the TBC on the interphase

thickness may also be arrived at using the virtual crystal extension of the DMM (VCDMM) that has been previously reported.[20] The virtual crystal approach, like the procedure employed here, accounts for the finite thickness of the interphase, but rather than treating the region as disordered, the model instead describes it as a hybridized crystalline solid [i.e., a virtual crystal (VC)] having an amalgam of properties from those of the substrate and the film. The TBC is subsequently predicted by scaling the interfacial conductance at the film/VC and VC/substrate junctions by the mean free path of the phonons as they propagate through the virtual crystal. While the technique is capable of accurately predicting the trend of TBC with interphase thickness, its use is limited due to the fact that it models a distinctly disordered region as a perfectly crystalline solid. Implicit in this assumption is that the thermal conductivity, and hence the mean free path that scales the TBC, must be quantified by approaching the interphase region as an alloyed crystalline solid in conjunction with Abeles original virtual crystal model.[38] Irrespective of the physical validity of the crystalline assumption, Abeles thermal conductivity model is constrained to temperatures above 300 K, thus bounding the capability of the VCDMM in its prediction of the TBC to a small region near room temperature where the effects of inelastic scattering remain diminutive.[19]

To examine if, unlike the VCDMM, the δ-DMM can accurately predict the temperature dependence of the TBC in the limit of dominant elastic scattering, an aluminum/sapphire (Al/Al_2O_3) system experimentally investigated by Swartz and Pohl[4] was evaluated in the temperature range of 5–150 K. This particular interface is chosen as significant disorder was induced through ion bombardment during synthesis, thereby ensuring the applicability of the present approach. In this range, three different methods were utilized to predict the thermal boundary conductance: the traditional DMM [Eq. (5)], the DMM in conjunction with electron/phonon coupling conductance $h_{e\text{-}ph}$, and finally the δ-DMM. For each prediction, material properties were acquired from Ref. 4 and, when applicable, the electron-phonon coupling factor for Al from Ref. 37. Temperatures above 150 K were not analyzed due both to the range of the experiment and the fact that the influence of inelastic scattering becomes more apparent at these levels.[32]

Unlike the Cr/Si interfaces that were examined above, the extent of disorder at the interface is not explicitly known in this instance. Consequently, D_{Int} is regarded as a free parameter in the evaluation of the δ-DMM with its value chosen through an optimization between the predicted and the experimentally derived values of the TBC. Using this methodology, the thickness of the interphase region, D_{Int}, was calculated to be 11 nm, a value that seems reasonable considering that the disorder induced by the bombardment was originally reported by Swartz and Pohl[4] to be of the order of 5–10 nm. Furthermore, throughout the entire temperature range, the δ-DMM is shown to be more capable of describing the TBC than either of the approaches based strictly on a crystalline solid (see Fig. 4). The enhanced predictions arise as a result of the more adept ability of the δ-DMM to account for the changes in the temperature dependent heat capacity and the thermal conductivity that occur in conjunction with the disorder. As a consequence of this finding, it may then be reasoned that reductions in the TBC are not only due to the finite thickness of an imperfect interface but also the disorder within the boundaries and the layers themselves that is implicit with this imperfection.

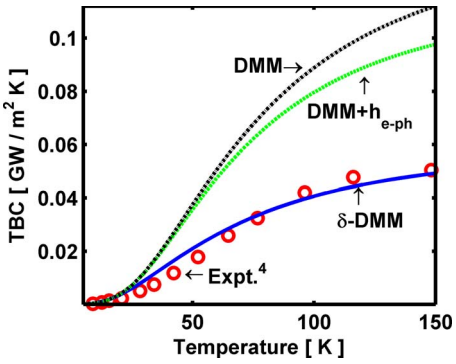

FIG. 4. (Color online) Thermal boundary conductance as a function of temperature for an Al/Al_2O_3 interface. Experimental values are obtained from Ref. 4 and are compared to the DMM in both its standard form as well as accounting for electron-phonon coupling resistance. The δ-DMM is evaluated assuming an 11 nm interphase thickness, D_{Int}, that is in line with the original assessment of the system and shows improved prediction over the crystalline based DMM models.

To probe the applicability of utilizing the interphase thickness as a free parameter in the analysis of a general system, a series of metal/substrate interfaces was examined at 300 K using the δ-DMM. In each case, the materials were selected to be similar in their vibrational spectra as only systems where the Debye temperatures of the film and substrate were within 50% of one another were investigated. This limitation is warranted in order to minimize the affect of inelastic scattering on the system as, again, this effect is not included in the present model.[14,19,32]

Using the δ-DMM, D_{int} was varied in order that the predictions were within ~1% of the experimentally obtained values of Stevens *et al.*[12] For each of the five examined systems, the predicted interphase thickness was found to have a value between 0.5 and 8.5 nm, as shown in Table I. These values are certainly plausible especially considering that Stevens *et al.*[12] commented that the interfaces employed

TABLE I. Comparison of TBC measurements for a series of material systems at room temperature with predictions using both the standard DMM and δ-DMM. To evaluate the applicability of assuming an interfacial thickness constant, D_{int} was scaled in the evaluation of the δ-DMM in order to equate the prediction with the experimental values. For each of the material systems, the predicted disordered region was between 0.5 and 8.5 nm. Such levels of disorder are within the bounds of expectation and indicate the applicability of the employed disordered approach. Material properties utilized in the simulations were acquired from Refs. 4, 40, and 41.

Materials (Film/Substrate)	Expt.[a] (GW/m^2 K)	DMM (GW/m^2 K)	δ-DMM (GW/m^2 K)	D_{Int} (nm)
Al/Si	0.12	0.55	0.12	8.5
Al/GaN	0.19	0.35	0.19	1.5
Cr/AlN	0.20	0.99	0.20	2.75
Cr/GaN	0.23	0.32	0.23	0.5

[a]Reference 39.

are "more than likely not abrupt junctions," thus lending further credence to the disordered approach in the limit of elastic scattering. It should not be reasoned, however, that the δ-DMM is then capable of predicting the extent of interfacial disorder. Rather, the obtained values of the interphase thickness are to be seen as unverifiable estimations that due to their reasonable magnitude seem to support the employed methodology.

The ramifications of considering disorder lead to additional parameters that must be considered during the design of systems having multiple interfaces. First, the perfection of the interface between two materials will, as has been stated previously, be a significant determinant in the TBC of the system as a whole.[4,16,20] In addition, the current approach highlights the fact that, in the limit of disorder, TBC is not only determined by the vibrational spectra but also the atomic densities of the solids involved [see Eqs. (13), (14), and (20)]. This fact may be of acute pertinence in the design of systems that contain materials that are nanoporous such as are being suggested for several thermal isolation applications.[39] Thus, it is not only the properties of the materials that will determine performance but also their morphology as well.

V. CONCLUSIONS

An extension of the DMM is demonstrated that accounts for disorder within both the interface itself as well as the constituent materials making up the boundary. By employing assumptions of scattering within disordered materials that modify the DOS and accounting for the length over which the interfacial disorder persists, the δ-DMM is shown to greatly improve predictions of the TBC without extreme computational rigor. In addition, the method also illustrates that the TBC is not solely determined by the properties of the materials themselves but also their morphology and the quality of their intersection.

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