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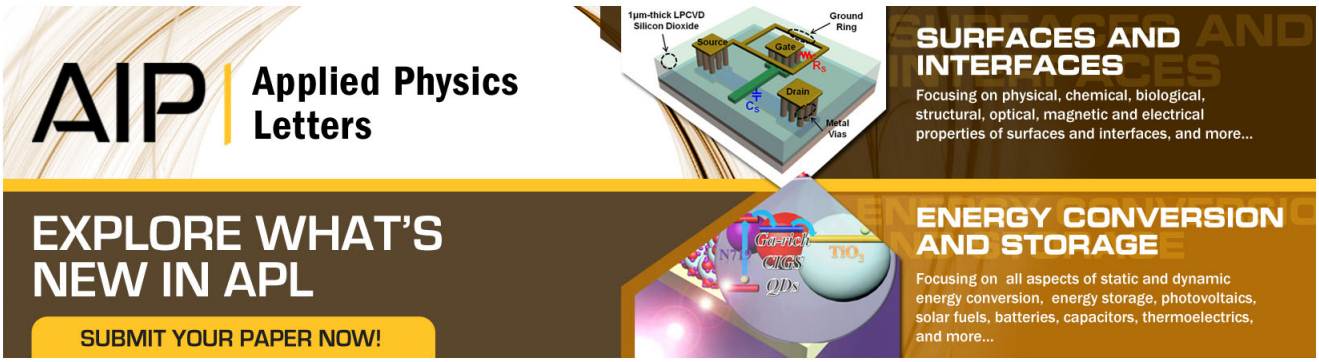

Role of interface disorder on thermal boundary conductance using a virtual crystal approach

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An analytical method is presented to estimate the effects of structural disorder on the thermal boundary conductance (TBC) between two materials. The current method is an extension of the diffuse mismatch model (DMM) where the interface is modeled as a virtual crystal of finite thickness with properties derived from those of the constituent materials. Using this approximation, the TBC for a series of chromium/silicon interfaces is modeled and shown to be within 18% of experimentally obtained values. The methodology improves upon the predictive capabilities of the DMM and allows for quick estimation of the impact of interface mixing on TBC. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2437685]

The transport of thermal energy across material interfaces is playing an ever increasing role in the response of thin-film and nanostructured devices. Thermal boundary conductance (TBC), which describes the efficiency of heat flow at material interfaces, is a concept which must be understood in order to control the thermal response of material systems such as superlattices, thermoelectrics, nanocomposites, and thermal interface materials. While much effort is under way to predict and understand TBC from atomistic calculations, a need still exists for more simplistic analytical calculations which capture salient aspects of the fundamental transport mechanisms leading to estimations of TBC. This is especially true when considering the impact of interface structural disorder on the thermal boundary conductance.

Most interfaces between thin films contain at least some degree of intermixing and are disorder dependent upon the processing methods employed. While atomistic modeling may capture details of the thermal energy transport across such disordered interfaces, it remains a daunting task to model all permutations of structural disorder which can be encountered in a real system. Thus, higher length scale analytical models which contain some statistically averaged quantities from the microstructure may provide a more tractable solution to such a problem.

The use of analytical models to estimate thermal boundary conductance between materials has frequently relied on the diffuse mismatch model (DMM).[1] This method assumes that when a phonon interacts with an interface it is diffusively scattered and hence transmission is completely dependent on the ratio of the densities of states between the two materials. Using this ratio, a transmission coefficient is calculated and the thermal boundary conductance is found by integrating over all frequencies using a Debye model to estimate the number of phonon carriers. Inherent in the diffuse scattering assumption is some degree of roughness or disorder at the interface. However, the nature of this roughness cannot be accounted for in any explicit or implicit manner using the DMM.

Approaching the Debye temperature, smaller phonon wavelengths are excited in the material, increasing the likelihood of multiple scattering events at an interface, which results in a decrease of the TBC. As the DMM predicts only a singular diffusive scattering event, it is not surprising that the model overpredicts TBC at noncryogenic temperatures in materials with disordered interfaces.[1–3] To improve upon the DMM, previous studies have largely focused on either using a more realistic density of states or investigating the extent to which phonons are indeed diffusively scattered.[3,4] While enhancements to the analytical approach of calculating TBC have been made through these efforts, there still exists a need to account for the extent of interface disorder on the estimation. This work seeks to account for these aspects of an interface to produce a method that quickly estimates the thermal boundary conductance while lending insight into the sensitivity of material systems to interface quality.

To account for interfacial disorder, a virtual crystal (VC) approximation for the interface will be made. In the virtual crystal approximation, the disordered region is replaced by a homogenized virtual crystal having effective properties based on the disordered medium.[5] Using this approach, transmission coefficients can be calculated on both sides of the homogenized crystal. The dependence of the thermal boundary conductance on the degree of disorder can then be examined by altering the composition of the virtual crystal. The thickness of the disordered interface is accounted through scaling of the conductance with respect to both the phonon mean free path in the virtual crystal and the thickness of the disordered region. Through these extensions, a model which can approximate the effects of interface structure on thermal boundary conductance is available.

The diffuse mismatch model calculates the thermal boundary conductance according to the following relation:

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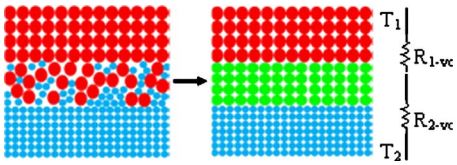

FIG. 1. (Color online) Schematic representation of the virtual crystal interface accompanied with associated heat resistance circuit. The interface region is replaced with a homogenized virtual crystal across which the phonon transmission coefficients are calculated, leading to TBC predictions.

$$h_{\text{int}} = \sum_j h_{1-2,j} = \sum_j \frac{1}{4} \int_0^{\omega_{\max}} \left(\frac{dN_{1,j}}{dT} \right) \hbar \omega \alpha_{1-2} c_{1,j} d\omega, \quad (1)$$

where $h_{1-2,j}$ is the TBC for a particular mode j, $\omega_{\max}$ is the Debye cutoff frequency, $\hbar$ is Planck's constant divided by 2π, $c_{1,j}$ is a given mode velocity, and N_j is the phonon distribution at the temperature and mode of interest based on the Debye theory. Of special note is the transmission coefficient α_{1-2} which quantifies the percentage of phonons able to forward scatter from side 1 to 2 and hence transmit thermal energy. The transmission coefficient is found to be a ratio of the phonon propagation speeds in each material:

$$\alpha_{1-2} = \frac{\sum_j c_{2,j}^{-2}}{\sum_j (c_{1,j}^{-2} + c_{2,j}^{-2})}. \quad (2)$$

In the diffuse mismatch model, it is assumed that the interface can cause only a single diffuse scattering event, neglecting any possibility that a phonon may be scattered multiple times. However, if there is a finite thickness of disorder at the interface, the assumption of a single scattering event may not be satisfied. In considering the interface between two materials, an interphase region of appreciable thickness with properties much different than those of the materials on either side may exist. Since the diffuse mismatch model cannot address this situation, it may be limited to applications of a perfect interface or where the interphase region is very small compared to the wavelength of the dominant heat carriers.

Modeling the interphase region is extremely complex as details of its structure and composition are not fully known. On the other hand, effective material properties which homogenize the interphase can yield some insight into the region's impact on TBC. Using such a method to estimate properties was first proposed by Abeles while investigating the thermal conductivity of mixed crystal alloys.[5] In this manner, prediction of the thermal conductivity in alloys such as SiGe was made possible over a wide range of compositions.

Through utilization of the virtual crystal approach, the interphase region is replaced by a virtual crystal, creating an interface with materials 1 and 2 as shown in Fig. 1. The total thermal boundary conductance is then found by finding the equivalent conductance of the two interface system.

$$h_{\text{int}} = \left[\left\{ \sum_j h_{1-\text{VC},j} \right\}^{-1} + \left\{ \sum_j h_{2-\text{VC},j} \right\}^{-1} \right]^{-1}. \quad (3)$$

To calculate the conductance between a material and the virtual crystal, Equation (1) is used with modifications to account for the effects of the virtual crystal. The only term which must be changed in this expression is the transmission ratio which will be modified by the presence of the virtual crystal. Equation (2) is now applied between material 1 and the virtual crystal as well as material 2 and the virtual crystal in order to calculate the TBC in Eq. (3).

To calculate the unknown phonon propagation speeds in the virtual crystal required in Eq. (2), the Debye theory may be used as proposed by Abeles.[5] However, this methodology requires the knowledge of the elastic constants of the virtual crystal. Near the interface, however, estimation of these constants is quite difficult due to the inherent randomness of the interface. As a consequence, it is assumed in this approach that the propagation can be modeled simply as a composite of the speeds of the host materials as shown below, where b_1 is the percentage by mass of material 1 in the VC:

$$c_{\text{VC},j} = b_1 c_{1,j} + (1 - b_1) c_{2,j}. \quad (4)$$

In considering the disorder analysis above, the depth of disorder has not been considered. To incorporate the effect of this interface thickness a dimensionless parameter, the depth factor δ, is introduced,

$$\delta_j = D/\Lambda_{\text{VC},j}. \quad (5)$$

In Eq. (5), D is the actual depth or thickness of the interface, while $\Lambda_{\text{VC},j}$ is the mean free path of the virtual crystal for a particular mode. This mean free path is calculated from the kinetic theory using the relation between thermal conductivity, average phonon speed, and specific heat in the bulk,

$$\Lambda_{\text{VC},j} = \frac{3K_{\text{VC}}}{C_{\text{VC}} c_{j,\text{VC}}}. \quad (6)$$

The calculation of the thermal conductivity in the virtual crystal is accomplished using the original approximation method by Abeles,[5] while all other parameters are estimated using the rule of mixtures as in Eq. (4). The depth factor relates the relative region of disorder to the dominant mean free path of the heat carriers. In this case, if $\delta > 1$, there is a possibility that the phonons can scatter multiple times in the interphase region. Thus, the boundary conductance should scale with this factor. The depth factor is used to modify Eq. (3) into the final form for evaluation of the thermal boundary conductance using the virtual crystal diffusive mismatch model (VCDMM).

$$h_{\text{int}} = \left[\left\{ \sum_j \frac{h_{1-\text{VC},j}}{\delta_j} \right\}^{-1} + \left\{ \sum_j \frac{h_{2-\text{VC},j}}{\delta_j} \right\}^{-1} \right]^{-1}. \quad (7)$$

In the case of metal-dielectric interfaces, the effect of electron-phonon coupling resistance should be accounted for by including the method of Majumdar and Reddy.[6]

To compare the current model to experimental data, the interfacial thickness D as well as the composition b of a disordered region must be known. Hopkins and Norris and Hopkins *et al.* examined the effect of these parameters on TBC using both Auger spectroscopy and a transient thermoreflectance technique for a series of chromium/silicon interfaces.[7,8] Using the reported interfacial thicknesses and deriving the virtual crystal composition from the given elemental concentration profiles, it is possible to compare the measured values of TBC to those predicted from the virtual crystal approach. These comparisons were carried out including electron-phonon coupling resistance using the value for chromium reported by Stevens *et al.*[9] and material properties obtained from Swartz and Pohl.[1] Shown in Table I is a com-

TABLE I. Comparison of thermal boundary conductance (TBC) calculated from the virtual crystal diffuse mismatch model (VCDMM) to measured values for several Cr/Si interfaces (Refs. 7 and 8). For each interface examined, the prediction is within 18% of the measured value.

Interfacial thickness (nm)	Virt. crystal composition (% Si)	Measured TBC ($GW/m^2 K$)	Virt. crystal DMM ($GW/m^2 K$)	DMM ($GW/m^2 K$)
9.5	0.54	0.178	0.147	0.855
14.8	0.67	0.113	0.118	0.855
11.5	0.66	0.139	0.146	0.855
10.1	0.48	0.15	0.131	0.855

parison of the virtual crystal model to the experimental data where in each case the VCDMM is within 18% of the measured values. From the table, it can be seen that increased interfacial thickness results in a reduction in TBC as the number of scattering events increases. These additional scattering mechanisms reduce the rate at which energy can be transported across the interface, causing a decrease in the TBC.

While the thickness of the interface region plays a strong role in the TBC, the disorder of that phase will also affect this parameter. This occurs as with increased disorder the number of scattering sites increases once again, limiting thermal transport. This effect is seen in Fig. 2 for an interface between Cr and Si of thickness of 9.5 nm for which the composition of the VC is allowed to vary from 10% to 90% Si. When the concentration is near one-half, and disorder is then maximized, TBC is minimized following the same qualitative trend of the effective thermal conductivity as predicted by the Abeles mixed crystal thermal conductivity model.

An extension of the diffuse mismatch model is presented for disordered interfaces. Utilizing a virtual crystal approximation for the interface, it is possible to estimate the thermal boundary conductance for interfaces with finite thickness and disorder. While the model qualitatively follows the trends of

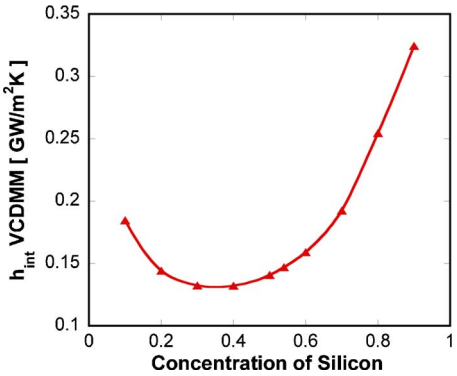

FIG. 2. (Color online) Thermal boundary conductance of a 9.5 nm Cr/Si interface as a function of Si content. The model shows a minimum thermal conductance occurring near 40% fraction of Si due to a peak in phonon scattering and minimization of thermal conductivity.

limited experimental data on this topic, additional effects must still be added to this approach to capture both the temperature dependence of the phonon mean free path and TBC. However, the current approach shows a simplistic method which captures physically motivated phenomena offering improvements over the diffuse mismatch model.

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