THERMAL TRANSPORT ACROSS TRANSFER PRINTED METAL-DIELECTRIC INTERFACES: INFLUENCE OF CONTACT MECHANICS AND NANOSCALE ENERGY TRANSPORT

BY

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THESIS

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ABSTRACT

Recent experiments suggest that the interfacial thermal conductance of transfer printed metal-dielectric interfaces is \(~45 \text{ MW/m}^2\text{K}\) at 300K, approaching that of interfaces formed using physical vapor deposition. In this work, we investigate this anomalous result using a combination of theoretical deformation mechanics and nanoscale thermal transport. We establish that the plastic deformation and the capillary forces lead to significantly large fractional areal coverage of \(~0.2\) which enhances the thermal conductance. At the microscopic transport scale, existing models that account for the electron-phonon non-equilibrium at the interface employ a phonon thermal conductivity that is difficult to estimate. We remove this difficulty by obtaining the conductance directly from the Bloch-Boltzmann-Peierls formula, describing the matrix element using a deformation potential that can be estimated from the electrical resistivity data. We report calculations up to 500 K to show that electron-phonon coupling is not a major contributor to the thermal resistance across metal-dielectric interfaces. Our analysis of the thermal conductance based on the consideration of both deformation mechanics and nanoscale thermal transport yields a conductance that is on the same order of magnitude \(~10 \text{ MW/m}^2\text{K}\) as the experimental data and partially follows the temperature trend. There remains a quantitative discrepancy between data and theory that is not explained through deformation of the interface alone. We suggest that capillary bridges formed in the small asperities may account for this discrepancy. A preliminary analysis shows this to be plausible based on available data. Our work advances the understanding of the role of electron-phonon coupling in limiting thermal transport near metal-dielectric interfaces and shows that, in terms of heat flow characteristics, metallic interconnects formed using transfer printing are comparable to ones formed using vapor deposition.
Acknowledgement

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1. INTRODUCTION

With the existing trend of miniaturization in electronic and optoelectronic devices, thermal management has become a critical issue with the ever increasing heat flux encountered in such devices. As the device dimensions shrink to ~10 nm, the thermal boundary resistance (TBR) at the interface becomes comparable to the intrinsic thermal resistance of the materials forming the device and cannot be ignored. As an example, a TBR of ~$10^{-8}$ m$^2$K/W for a GaN/Si field effect transistor dominates the intrinsic resistance for characteristic dimensions less than a few µm.$^1$ Hence, an accurate prediction of TBR is a critical aspect for devising thermal management strategies for micro-devices.

The TBR results in a temperature drop ($\Delta T$) at the interface which is related to heat flux ($q$) by:

$$q = G \times \Delta T$$

(1.1)

Here, $G$ is the interfacial boundary conductance, the inverse of TBR. TBR has origin in two distinct effects. The first one is a macroscopic component that arises due to the geometrical constriction of the actual contact area, $A_c$ compared to the nominal area, $A_n$ and is commonly referred to as the thermal contact resistance. The formulation of macroscopic resistance is based on Maxwell’s formula$^2$. Maxwell’s expression was originally derived for a constriction of circular geometry but it has been subsequently modified for arbitrary geometries.$^3$ The modified Maxwell’s expression considers only the contact load due to elastic/plastic deformation at the contact spot. However, when the dimensions of the contact spot are on the order of ~1 nm, the adhesive Van Der Waals forces and the capillary forces due to water bridges at the asperities can
alter the deformation zone geometry and hence, the macroscopic resistance. In this work, we address this issue and take both, the adhesive and the capillary forces into account for calculating the macroscopic resistance.

TBR also originates due to a microscopic effect which was first observed for an interface of solids with liquid helium by Kapitza\textsuperscript{4} in 1941. It was first explained by Khalatnikov using the Acoustic Mismatch Model (AMM)\textsuperscript{4} which was based on phonon reflection and transmission at the interface. However, the assumption of specular scattering of phonons at the interface in the AMM breaks down when the phonon wavelength becomes comparable to the surface roughness of the interface. This is particularly true for phonons near the room temperature and it lead Swartz and Pohl to propose the Diffuse Mismatch Model (DMM).\textsuperscript{4} DMM assumes diffuse scattering of phonons at the interface and that the subsequent transmission is based on the density of states of phonons on both sides. The DMM has been widely used to calculate the thermal conductance for dielectric-dielectric interfaces which matches reasonably well with experiments. However, experiments on metal-dielectric interfaces are generally in poor agreement with DMM values.\textsuperscript{5,6} The situation is different from dielectric substrates in that the majority heat carriers in metal are electrons. So, the pathway for energy transport is from the electrons in metal to the phonons in metal and then from the phonons in metal to the phonons in dielectric.\textsuperscript{7} Due to the non-equilibrium between electrons and phonons in the metal, there is an additional thermal resistance to the heat flow in series with the phonon-phonon resistance across the interface. The leading model\textsuperscript{7} gives the value of the thermal conductance for this component as \( G_{e-ph} = \sqrt{Gk_p} \) where \( G \) is the electron cooling rate and \( k_p \) is the phonon thermal conductivity of the metal. The value of \( G_{e-ph} \) depends upon the phonon thermal conductivity of the metal which is hard to extract from the total thermal conductivity data due to the dominance of electron
thermal conductivity. Also, the temperature dependence of $G_{e-ph}$ is analyzed under the assumption that $G$ is independent of temperature and $k_p \sim T^{-1}$. This gives an empirical expression: $G_{e-ph} = \psi / \sqrt{T}$, where the empirical factor is obtained by fitting the experimental data on thermal conductance after accounting for the DMM value. This procedure has shortcomings in that, first, the inverse temperature dependence of phonon thermal conductivity is only valid for temperatures comparable to the Debye temperature of the metal. At low temperatures, the phonon thermal conductivity goes as the third power of temperature. A more accurate but less convenient way would be to express $G_{e-ph}$ using empirical relations for different temperature ranges and obtaining the empirical factors separately for each range. Second, in this model it is assumed that $G_{e-ph}$ and DMM conductance are the only contributing factors to the net thermal conductance. However, interfacial defects, mixing and substrate damage lead to lowering of thermal conductance which is unaccounted for. Clearly, there is need for a better model which can predict $G_{e-ph}$ independent of thermal conductance measurements. In this work, we address this issue by developing a new model using electron-phonon scattering theory and the use of more reliable electrical resistivity data to extract the empirical parameter. Our work on electron-phonon thermal conductance will provide an insight into the energy transport across metal-dielectric interfaces in existing and upcoming technologies like MOSFETs and spintronics.

The macroscopic component dominates the overall resistance when the size of the constriction is very large as compared to the mean free path of the energy carriers. This is typically the case for bulk machined and mechanically pressed contacts where the surface roughness is $\sim$1-100 $\mu$m. In the opposite limit, as for the interfaces formed by physical vapor deposition, the microscopic component dominates. However, for transfer printed interfaces, both
the effects gain relative importance. The transfer printed metal film originally formed by vapor deposition has a surface roughness of ~0.1-1 nm which is orders of magnitude lower than that for bulk-machined interfaces but it still does not form an atomically smooth interface as in the case of vapor deposited interfaces. As a result, both components should be taken into consideration while calculating the thermal conductance.

The interfacial thermal conductance of transfer printed objects is a relatively less explored field. Recent experiments\textsuperscript{10} show that the interfacial thermal conductance for transfer printed gold films on silicon dioxide substrate is surprisingly high, \(\sim 40\ \text{MW/m}^2\text{K}\) in the temperature range 300-600 K. This is surprising since the value far exceeds the typical thermal conductance of bulk-machined and mechanically pressed interfaces (\(\sim 0.1\ \text{MW/m}^2\text{K}\)) and approaches that of interfaces formed by vapor deposition under vacuum (\(\sim 10-700\ \text{MW/m}^2\text{K}\)). Significant fractional area coverage was hypothesized as the reason for high thermal conductance; however it was not backed by actual calculations. In this work, we theoretically calculate the fractional area of contact using deformation mechanics and relate it to the macroscopic thermal conductance. We focus on the role of attractive adhesive forces and capillary forces in augmenting the actual area of contact. The results of our work will advance the understanding of deformation mechanics and electron-phonon coupling at metal-dielectric interfaces that will aid in devising thermal management strategies for transfer-printing based technologies like stretchable electronics.\textsuperscript{11}

1.1 Structure of the thesis

This thesis is organized in the following manner: In Chapter 2, a theoretical framework for calculating the macroscopic thermal conductance for transfer printed interfaces is presented. The effect of contact mechanics on actual contact area which in turn affects interfacial thermal...
conductance is shown. We present a way to incorporate the adhesion forces and the capillary forces to better estimate the contact area and the macroscopic thermal conductance. In chapter 3, we present a new model to calculate the electron-phonon thermal conductance. We perform calculations for a variety of metals across a wide range of temperature and show that our model is better applicable as compared to the previous leading model from literature. We further answer the question that whether electron-phonon thermal conductance can be ignored as compared to DMM values for typical metal-dielectric interfaces. In chapter 4, we combine the macroscopic and microscopic components of interfacial thermal conductance to calculate the net thermal conductance. We then compare the calculations to the existing experimental studies. Finally in Chapter 5, the summary of the results from Chapters 2, 3 and 4 is presented and the conclusions of these results along with the scope of future work are discussed.
2. MACROSCOPIC COMPONENT OF THERMAL CONDUCTANCE

A typical transfer printed interface is shown in figure 2.1. Both the metal and dielectric objects have asperities on their surfaces. If the transfer printing is done in ambient air, water from ambient air can be present on the asperities and can form capillary bridges as shown in figure 2.1. Various modes of heat transport across such an interface are: Solid-to-solid conduction through asperities in contact, heat conduction through the air gap, near field electromagnetic (EM) radiation and conduction through the capillary bridges. Recent experiments on Au-SiO₂ and Au-Si interfaces¹² revealed a thermal conductance, \( G \approx 40 \text{ MW/m}^2\text{K} \). The upper limit on heat transport by near field EM radiation and air conduction is \( \approx 0.1 \text{ MW/m}^2\text{K} \) which is two orders of magnitude smaller than the experimentally observed result.¹²,¹³ Hence, we ignore these two components in the subsequent analysis and now focus on solid-to-solid conduction.

![Figure 2.1: Various modes of heat transport across a transfer-printed metal-dielectric interface. Yellow color represents the metal, green color represents the dielectric and blue color represents the water bridges.](image)
2.1 Contact Mechanics

For an individual asperity in contact with a flat surface, the macroscopic interfacial thermal conductance $G_i$ is given by\(^2\)

$$G_i = A_i^{-1} \left[ \frac{\varphi_i}{4k_1k_2a_i / (k_1 + k_2)} \right]^{-1} \quad (2.1)$$

where $A_i$ is the area of the adiabatic flux tube, $\varphi_i$ is a geometrical factor given as $\varphi_i = (1 - a_i/b_i)^{3/2}$, $b_i$ is the flux tube diameter for the contact, $a_i$ is the radius of the contact area, $k_1$ and $k_2$ are the thermal conductivities of the contacting materials. We note that the area $A_i$ equals $\pi b_i^2$. Assuming the same flux tube diameter for all contacting asperities, i.e. $b_i = b$, the total macroscopic interfacial thermal conductance $G_{macroscopic}$ due to all contacts is

$$G_{macroscopic} = \eta A \int_d^\infty G_i \phi(z) dz = \left[ \eta \int_d^\infty \frac{1}{(1-a_i/b)^{3/2}} / 2k_H a_i \phi(z) dz \right]^{-1} \quad (2.2)$$

where $k_H$ is the harmonic mean of thermal conductivities given by $k_H = 2k_1k_2 / (k_1 + k_2)$, $\eta$ is the areal density of asperities, $d$ is the separation between the surfaces and $\phi(z)$ is the height distribution function for asperities. The average flux tube diameter $b$ is then

$$b = \pi \eta \int_d^\infty \phi(z) dz]^{-1/2}$$

where $\eta \int_d^\infty \phi(z) dz$ is the density of contacting asperities. We subsequently use deformation mechanics to obtain $a_i$. If we further assume the contact radius of each asperity $a_i$ to be a constant equal to $a$, Eq. (2.2) simplifies to

$$G_{macroscopic} = \left[ \frac{(1 - A^{1/2})^{3/2}}{2k_H a} \right]^{-1} \quad (2.3)$$

\[7\]
where $A$ is the ratio of actual area $A_c$ to nominal area $A_n$, and $a$ is given by $a = A^{1/2} \left[ \pi \eta \int_{d}^{\infty} \phi(z)dz \right]^{-1/2}$. We find that the error between the prediction from the simplified expression above and that from the more accurate expression in Eq. (2.2) is less than 1% when surface roughness is < 1 nm as is the case here.

Consideration of surface roughness is an important aspect in estimating the actual area of contact. Even for a “smooth” surface, there are asperities of random heights and radii. Contact between surfaces happens at asperities larger than the separation gap. Quantitative evaluation of the actual contact area remains challenging despite decades of research. The seminal model was developed by Greenwood and Williamson (GW model). $^{15}$ The GW model mainly predicts the contact area and the contact load based on elastic deformation of contacting surfaces. The central idea is to reduce the problem of two contacting rough surfaces to that of contact between a single rough surface and a perfectly flat surface as shown in Fig. 2.2. Asperities are assumed to be spherical with radius $R$, at height $z$ from the reference plane (see Fig. 2.2) and are assumed to follow a known probability distribution function $\phi(z)$. The model further assumes that the elastic deformation of an individual asperity follows the Hertz theory $^{16}$ while the distribution of asperity heights is Gaussian.

When the interference, $\omega$, defined as the difference between asperity height and the surface separation based on a reference plane defined by the mean of asperity heights, exceeds the critical interference, $\omega_c$, deformation is no longer strictly elastic. There are two notable modifications to the GW model that consider plastic deformation. The first is based on the work of Chang et al. (CEB model)$^{17}$, and the second on the work of Kogut and Etsion (KE model)$^{18}$. The CEB model models plastic deformation under the simplified assumptions of uniform
average pressure of each contacting asperity and conservation of its volume. While this elastic-plastic model shows improvements over the GW model, the CEB model is inaccurate for highly plastic or fully plastic deformation due to its simplified assumptions. More recently, Kogut and Etsion\textsuperscript{18} extended the CEB model to include finite element analysis (FEA)\textsuperscript{19}, enabling force and area calculations under fully plastic contact. We employ this model in the present work and later show that this yields more physically plausible results than the other two models in the case of deformation during transfer printing.

Figure 2.2: Schematic illustrating various parameters used in deformation mechanics model.

2.2 Modeling of contact area and contact load

The KE model extends the classical Hertzian solution from elastic contact to the case of fully plastic contact using accurate finite element analysis based on constitutive laws for all deformation modes. For the KE model, the dimensionless contact area, $A^*$ and the contact load, $P^*$ are given by
\[
A'(d^*) = \frac{A}{A_n} = \pi \eta R \sigma \omega_c^* \left( I^1 + 0.93 \int_{d' + \theta_0^*}^{d'+6\theta_0^*} I^{1.136} + 0.94 \int_{d' + 6\theta_0^*}^{d'+10\theta_0^*} I^{1.146} + 2 \int_{d' + 10\theta_0^*}^{\infty} I^1 \right)
\]

(2.4)

\[
P'(d^*) = \frac{P}{A_n H} = \frac{2}{3} \pi \eta R \sigma K \omega_c^* \left( I^{1.5} + 1.03 \int_{d' + \theta_0^*}^{d'+6\theta_0^*} I^{1.425} + 1.4 \int_{d' + 6\theta_0^*}^{d'+10\theta_0^*} I^{1.263} + \frac{3}{K} \int_{d' + 10\theta_0^*}^{\infty} I^1 \right)
\]

(2.5)

where the general form of the integrand is, 

\[
I^\alpha = \left( \frac{z^* - d^*}{\omega_c^*} \right)^\alpha \phi^*(z^*) dz^*
\]

and \(d\) is the separation based on asperity heights, \(A_n\) is the nominal contact area, \(\eta\) is the areal density of asperities, \(R\) is the asperity radius of curvature, \(\sigma\) is the standard deviation of surface heights, \(H\) is the hardness of the softer material, \(K\) is the hardness coefficient related to the Poisson’s ratio of the softer material given as \(K = 0.454 + 0.41\nu\), and \(z\) is the height of asperity from the mean of asperity heights. The normalized Gaussian distribution function \(\phi^*(z^*)\) is defined as \(\phi^*(z^*) = \frac{\sigma^*}{(2\pi)^{1/2}\sigma} \exp[-0.5(\sigma z^*/\sigma_S)^2]\) where \(\sigma_S\) is the standard deviation of asperity heights. The superscript * denotes dimensionless values while all the lengths are non-dimensionalized with respect to \(\sigma\).

We first calculate the contact load and contact area for transfer printed Au-SiO\(_2\) interface as a function of the separation distance. We include predictions from the GW and CEB models for comparison but do not employ these in the subsequent analysis. The models require three input parameters: the asperity radius of curvature \(R\), the standard deviation of surface heights \(\sigma\), and the areal density of asperities \(\eta\). These are 0.136 \(\mu\)m, 0.382 nm, and 640 \(\mu\)m\(^2\) respectively. We determine these from AFM surface profiles\(^{12}\) using equations suggested by\(^{20}\):
\[ R = \frac{3\sqrt{\pi}}{8\left\langle \left(\frac{d^2 \zeta}{dx^2}\right)^2\right\rangle^{1/2}} \]  \hspace{1cm} (2.6)

\[ \eta = \frac{\left\langle \frac{d^2 \zeta}{dx^2} \right\rangle}{\frac{\left\langle d\zeta / dx \right\rangle}{6\pi\sqrt{3}}} \]  \hspace{1cm} (2.7)

\[ \sigma = \left\langle \zeta^2 \right\rangle^{1/2} \]  \hspace{1cm} (2.8)

where \( \zeta \) is the measured height, \( dx \) is the distance between two adjacent measurements, and the operator \( \left\langle \right\rangle \) denotes an arithmetic average.

### 2.3 Modeling of adhesion and capillary forces

When two surfaces are in contact, four forces, namely the contact load \( P \), the external force \( F_{ex} \), the adhesion force \( F_s \), and the capillary force \( F_{cap} \) balance each other as \( P = F_{ex} + F_s + F_{cap} \) as shown in figure 2.3. When an external force is absent, the contact load must balance the adhesion force and the capillary force. While the original work from Greenwood and Williamson does not include consideration of adhesion forces, Johnson et al.\textsuperscript{21} and Derjaguin et al.\textsuperscript{22} proposed two different models, the JKR model and the DMT model respectively, to consider adhesion between an elastic sphere and a flat surface. The major differences between these models are that the JKR model assumes that the adhesion forces lie within the contact area and increase the area of contact, while the DMT model predicts that the adhesion forces are exerted only outside the contact area and have no effect on the deformation of the sphere. Previous work\textsuperscript{23} has concluded that the JKR and the DMT models are both limiting cases of a general solution: The JKR model holds for materials with low elastic modulus while the DMT model holds for materials with high elastic modulus. Therefore, the DMT model is more suitable for
metallic contacts. Here, we apply the DMT model in the subsequent calculations and include a modification introduced by Muller et al.\textsuperscript{24} The original DMT model only considered a sphere in contact with a flat surface. Instead, Muller et al.\textsuperscript{24} assumed the dominant adhesion force to be the van der Waals force and the total intermolecular force to follow the Lennard-Jones surface pressure given by

\[ p(z) = \frac{8 \Delta \gamma}{3 \varepsilon} \left[ \left( \frac{\varepsilon}{z} \right)^3 - \left( \frac{\varepsilon}{z} \right)^9 \right] \]  

(2.9)

where \( \gamma \) is the energy of adhesion, \( \varepsilon \) is the intermolecular distance ranging between 0.3 and 0.5 nm, and \( z \) is the separation of the contacting sphere and the flat surface outside the contact area. Using this, we can write the dimensionless adhesion force \( F_z^\ast \) predicted by the KE model as

\[
F_z^\ast (d^\ast) = \frac{F_z}{A^\ast H} = 2\pi \eta R \frac{\Delta \gamma}{H} \left( \int_{d^\ast}^{d^\ast + \theta_{c}^\ast} J_{nc}^{d^\ast} + 0.98 \int_{d^\ast}^{d^\ast + \theta_{c}^\ast} J_{0.298}^{0.29} + 0.79 \int_{d^\ast}^{d^\ast + \theta_{c}^\ast} J_{0.356}^{0.321} + 1.19 \int_{d^\ast}^{d^\ast + \theta_{c}^\ast} J_{0.093}^{0.332} \right).
\]

(2.10)

where the integrand for the contribution of the non-contacting asperities is

\[
J_{nc} = \frac{4}{3} \left[ \left( \frac{\varepsilon^\ast}{d^\ast - z^\ast} \right)^2 - 0.25 \left( \frac{\varepsilon^\ast}{d^\ast - z^\ast} \right)^8 \right]
\]

and the general form of the integrand for the contribution of the contacting asperities is

\[
J_{\beta} = \left( \frac{z^\ast - d^\ast}{\omega_c^\ast} \right)^\alpha \left( \frac{\varepsilon^\ast}{\omega_c^\ast} \right)^\beta \phi^\ast (z^\ast) dz^\ast.
\]

For the consideration of capillary forces, here we assume a monolayer of water with thickness, \( t_w = 0.278 \) nm at the surface. The formation of water droplets on the surface follows the Kelvin equation.\textsuperscript{25} The resulting force applied to an individual asperity is\textsuperscript{26}
\[ F_{\text{cap}} = 2\pi R\gamma_w (1 + \cos \theta_c) \left(1 + \frac{d - z}{t_w}\right)^{-\kappa} \]  \hspace{1cm} (2.11)

where \( \gamma_w \) is the surface tension of water, \( \theta_c \) is the contact angle between water and the contacting asperity, and \( \kappa = 1 \) when \( (d - z) > 0 \) or zero otherwise. Therefore, we write the total capillary force as

\[ F_{\text{cap}}(d) = 2\pi R\gamma_w (1 + \cos \theta_c) \eta A_n \times \left\{ \int_{d-t_w}^{d} \left(1 + \frac{d - z}{t_w}\right)^{-1} \phi(z)dz + \int_{d}^{\infty} \phi(z)dz \right\} \]  \hspace{1cm} (2.12)

Figure 2.3: Schematic illustrating various forces acting along a transfer printed interface. P is the contact load, \( F_s \) is the adhesion load, \( F_{\text{cap}} \) is the capillary force and \( F_{\text{ext}} \) is the external force.

For ease of calculation, we assume that the capillary forces and the intermolecular forces are independent and the water layer does not influence the surface adhesion energy in Eq. (2.10). Furthermore, we assume the contact angle, \( \theta_c \), to be 80° in all cases. Surface conditions such as surface contaminants and oxide formation affect the wettability of metals, resulting in drastic changes in contact angles. Au shows a surprisingly wide range of contact angles between 0-
While our choice of contact angle is nominal, we note that using a contact angle of zero increases both the contact area and the thermal conductance by ~23%.

Figure 2.4 shows the contact load and the adhesion forces as a function of separation from the mean of asperity heights. Wet adhesion refers to the inclusion of the capillary force and dry adhesion refers to the exclusion of this force in the calculations. We show the calculations for the CEB model for comparison. The KE model predicts the force balance at a separation of \( \sim (- \sigma) \) whereas for CEB model the balance occurs at \( \sim (-4 \sigma) \). Considering the highly malleable characteristics of Au, the prediction of the KE model is reasonable while that of the CEB model is clearly not. For both models, the contact load increases as the mean separation decreases, while the rate of increase slows around 0.35 nm. The latter is comparable to the standard deviation of surface heights, \( \sigma \). For the KE model, both wet and the dry adhesion are maximum around a mean separation of zero, and decrease when the separation decreases further. As given in Eq. (2.10), the fully plastic contact becomes more significant as the separation becomes smaller. However, the effect of adhesion force due to the fully plastic contact is smaller than forces in other deformation modes, and thus overall, the adhesion forces decline.
Figure 2.4. Comparisons between the CEB (solid lines) and the KE (dashed lines) models in the calculations of the contact load $P$ and the adhesion force $F_s$. Dry adhesion excludes water capillary forces while wet adhesion includes this. The force balance between the contact load and the adhesion forces happens at $\sim(-\sigma)$ for the KE model and at $\sim(-4\sigma)$ for the CEB model.

We next investigate the change in the contact area with increasing temperature. We obtain temperature dependent parameters for Au and SiO$_2$ surfaces from the literature.$^{31-33}$ The main temperature dependence is in the Young’s modulus and hardness of Au that decrease with increasing temperature, resulting in greater deformation of contacting asperities. Figure 2.5 shows the calculated contact area as a function of temperature for the Au-SiO$_2$ interface. In the case of dry adhesion, the contact area increases marginally from ~10% to ~11% as temperature increases from 300K to 600K due to increased deformation as discussed above. However in the case of wet adhesion, the dominant adhesion force due to surface tension decreases as
temperature increases which leads to lesser augmentation of contact area due to capillary forces at higher temperatures. This results in a steep decrease in the contact area from ~21% to ~13% for the same temperature change of 300K.

Figure 2.5. The actual contact area between Au and SiO₂ surfaces as a function of temperature. For dry adhesion, the contact area increases due to decrease in the Young’s modulus $E$ and hardness $H$ of the softer material, Au in this case. In wet adhesion, the area decreases since the surface tension of water decreases as temperature increases.

A key parameter affecting the area of contact is the plasticity index, $\psi = \frac{2E}{\pi KH} (\sigma_s R^{-1})^{1/2}$, that indicates the degree of elastic and plastic deformation. According to Kogut and Etsion¹⁸, deformation transitions from elastic to elastic-plastic around $\psi \sim \sqrt{2}$ and asperities start reaching fully plastic deformations only if $\psi > 6$. In order to investigate the influence of the plasticity index on the actual contact area, we examined eight different metals as shown in table 2.1.
<table>
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<th>Al</th>
<th>Au</th>
<th>Ag</th>
<th>Cu</th>
<th>Pt</th>
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<td>0.549</td>
<td>0.638</td>
<td>1.06</td>
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<tr>
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<td>3.08</td>
<td>0.971</td>
</tr>
</tbody>
</table>

Table 2.1. Material properties of various metals at 300K. The plasticity index Ψ is determined by the mechanical properties of contacting materials and the surface roughness parameters, and indicates the degree of elastic and plastic deformation in each case.

Assuming that the energy of adhesion between the metal and the nonmetal arises from London dispersion forces, the range for adhesion energy of most metals is quite narrow, within 0.06-0.1 Jm⁻². In these calculations, we assumed the energy of adhesion to be constant at γ=0.1 Jm⁻². Figure 2.6 shows resulting contact areas for these materials, assuming roughness similar to that for Au. The actual contact area is proportional to the plasticity index with the trend slowing at Ψ~8. This trend agrees well with previous results. Figure 2.6 also shows the contact area under wet adhesion. The contact area increases ~2 times compared to dry adhesion, for most metals. From this we infer that it is the combined effect of plastic deformation and capillary forces which leads to significant enhancement of contact area. We show later that the interfacial thermal conductance for different transfer-printed metallic films is strongly dependent on the plasticity index.
Figure 2.6. Predictions of the actual contact area for different metals showing the strong dependency of the contact area on the plasticity index. Blue dots represent wet adhesion and red dots represent dry adhesion. The contact area linearly increases when the index is low ($\psi < 6$), but the trend slows once the index becomes larger than 8.
3. MICROSCOPIC COMPONENT OF THERMAL CONDUCTANCE

3.1 Diffuse Mismatch Model (DMM)

Thermal resistance at the interface of two solids is observed even for atomically perfect interfaces. The origin of this resistance, named as the microscopic component here, is different from macroscopic component. Two models, namely, Acoustic Mismatch Model (AMM) and Diffuse Mismatch Model (DMM) have been proposed to explain the origin and calculate this thermal resistance. In AMM, the phonons are treated as waves, the solids are treated as continua, the interface is treated as a plane and the phonon propagation is governed by continuum acoustics. The interfacial thermal resistance is then explained based on different acoustic impedances of the two solids which depends on properties like density and speed of sound in the solid. The wave picture of phonon is not valid at room temperature for typical vapor deposited interfaces, when the wavelength of thermally excited phonons (~0.1 nm) is comparable to the interfacial roughness. For such cases, DMM is better applicable.

The idea behind DMM is that the phonons are scattered diffusely at the interface and as a result phonon wave vector and polarization is completely randomized. If $a$, $i$ and $j$ represent the probability of transmission of a phonon across the interface, index for medium from which the phonon transmits ($i=1, 2$) and polarization of phonon ($j=1, 2, 3$) then because of randomization of polarization and wave vector

$$\alpha_{i,j}(\omega(\vec{k})) = \alpha_i(\omega) \tag{3.1}$$

where $\omega$ is the frequency and $k$ is the wave vector of phonon. Also since a phonon is randomized after scattering, there is no way of telling whether a phonon in a particular medium has arrived
after transmission from the other medium or after reflection at the interface in to the same medium. This gives $\alpha_1 = 1 - \alpha_2, \alpha_2 = 1 - \alpha_1$.

To calculate $\alpha$, principle of detailed balance is applied. The number of phonons of frequency $\omega$ leaving side 1 per unit time per unit area is

$$\sum \int \int N_{1,j}(\omega, T) \times \alpha_1(\omega) \times c_{1,j} \cos \theta \times \sin \theta d\theta d\phi$$

where $N_{1,j}$ is the phonon density of states per unit solid angle for $j$th polarization and $c_{1,j}$ is speed of sound for that particular polarization. Assuming that the solids are isotropic Debye solids we get

$$N_{1,j} = \frac{\omega^2}{8\pi^3 c_{1,j}^3 \left[ \exp \left( \frac{\hbar \omega}{k_B T} \right) - 1 \right]}.$$ 

Similarly the number of phonons of frequency $\omega$ leaving side 2 per unit time per unit area is given by

$$\sum \int \int N_{2,j}(\omega, T) \times \alpha_2(\omega) \times c_{1,j} \cos \theta \times \sin \theta d\theta d\phi.$$ 

According to the principle of detailed balance the number of phonons of a particular state leaving a particular side must be equal to the number of phonons in the same state returning from other side at thermal equilibrium which means that the number of phonons leaving side 1 and side 2 must be equal. Equating the expressions and using the relationship between transmission probabilities we get

$$\alpha_1(\omega) = \frac{\sum_i c_{2,j}^{-2}}{\sum_j \sum_i c_{1,j}^{-2}} = \frac{2c_{2,j}^{-2} + c_{2,j}^{-2}}{2c_{1,j}^{-2} + c_{1,j}^{-2} + 2c_{2,j}^{-2} + c_{2,j}^{-2}} \quad (3.2)$$

where $l$ and $t$ represent the longitudinal and transverse modes respectively.

The net heat flow rate from material 1 to 2 is then the difference between the gross heat flow from side 1 to side 2, when side 1 is at temperature $T_1$ and the gross heat flow from side 2 to side 1 when side 2 is at temperature $T_2$. Using the principle of detailed balance, gross heat flow from
side 2 to side 1 when side 2 is at temperature $T_2$ is same as the gross heat flow from side 1 to side 2 when side 1 is at temperature $T_2$. From this, the net heat flow rate from side 1 to side 2 is given by:

$$\frac{dQ_{\text{net}}}{dt} = \frac{dQ_{1\to2}(T_1)}{dt} - \frac{dQ_{1\to2}(T_2)}{dt}$$

(3.3)

$$\frac{dQ_{1\to2}(T)}{Adt} = \sum_j \int_0^{\omega_1,\text{Debye}} \int_0^{2\pi} \int_0^{\pi/2} N_{1,j} \times \hbar \omega \times \alpha_i \times c_{i,j} \cos \theta \times \sin \theta \, d\theta \, d\phi \, d\omega$$

where $\omega_{1,\text{Debye}}$ is the Debye cut-off frequency of material 1 and $A$ is the area of cross section. Net heat flow is related to thermal conductance by $\frac{dQ_{\text{net}}}{dt} = G_{\text{ph-ph}} \times A \times (T_1 - T_2)$ where $G_{\text{ph-ph}}$ is the interfacial thermal conductance. The subscript ph-ph represents the interaction between phonons on both sides and its relevance will become clear in the next section. From the above expression, $G_{\text{ph-ph}}$ can be calculated in the limit $T_1 - T_2 \to 0$ as

$$G_{\text{ph-ph}} = \sum_j 2\pi c_{1,j} \tau_{1,j} \int_0^{\omega_1,\text{Debye}} \hbar \omega \frac{dN_{1,j}}{dT} \, d\omega$$

(3.4)

$$\tau_{1,j} = \int_0^{\pi/2} \alpha_i \cos \theta \, \sin \theta \, d\theta$$

The above discussion holds good for atomically perfect interfaces. The transmission coefficient is further modified because of scattering at irregular shaped asperities. The dominant wavelength of thermally excited phonons in Au, $\lambda_D$ is $\sim$0.1 nm when estimated from the Debye theory as $\lambda_D = 2\pi \hbar v/(k_b T)$, where $v$ is the Debye speed. As calculated in previous chapter for Au, $R$ is 136 nm and the constriction radius, $a$, for dry and wet adhesion are 8 nm and 12 nm respectively. Since $R > \lambda_D$ and $a > \lambda_D$, there is an additional geometrical scattering. The effect of geometric scattering is taken into account by multiplying the transmission coefficient by
Following the similar steps for calculation we get the relationship: 
\[ G_{ph-ph, \text{ballistic}} = \frac{2}{3} G_{ph-ph} \].

However, the modified value gives a lower bound of the interfacial thermal conductance because the condition for geometric scattering takes place only at certain locations.

### 3.2 Electron-Phonon thermal Conductance

DMM values match the experimental values reasonably well for interfaces formed by contact of dielectric surfaces when the dominant heat carriers are phonons. Intuitively, DMM should not hold good for metal-dielectric interfaces as the dominant heat carriers in metal are electrons and heat needs to be transported from electrons in metal to phonons in dielectric. Experimentally, this has been confirmed and measurements of the thermal conductance at metal-dielectric interfaces are generally in poor agreement with DMM values. Some of the earlier thermal conductance measurements\(^5,6\) revealed that DMM overestimates or underestimates the experimental values depending on the mismatch in the Debye temperatures of the metal and the dielectric. When the Debye temperature ratio of metal to dielectric is greater than 0.4, DMM overestimates the conductance.\(^6\) The various mechanisms proposed in literature for energy transport are shown in figure 3.1. [A] represents the transfer of energy from electrons in metal to phonons in metal.\(^7\) [B] represents the transfer of energy between phonons in metal to phonons in dielectric via elastic process described by DMM.\(^4\) [C] represents the transfer of energy between phonons via inelastic processes.\(^35\) [D] represents the transfer of energy between phonons in metal and dielectric via the interfacial mixing zone\(^36\) and [E] represents the transfer of energy between the electrons in metal to phonons in dielectric directly.\(^37\) For the cases when DMM under-predicts the experimental value, pathway [C] provides an explanation. For this case when the Debye temperature of metal
and dielectric are widely mismatched, two phonons of lower frequency on metal side can combine and create a phonon of higher frequency on dielectric side. Energy conservation dictates:

\[ h\omega_{metal} + h\omega_{metal} = h\omega_{dielectric} \]

\[ \Rightarrow \omega_{dielectric} = 2\omega_{metal} \]

\[ \Rightarrow (\omega_{dielectric})_{\text{max}} = 2\omega_{\text{Debye,metal}} \]  

(3.5)

As long as \((\omega_{dielectric})_{\text{max}}\) is less than Debye cut-off frequency of dielectric, the three phonon process can occur. The theoretical temperature trend of this mechanism has been verified experimentally for widely mismatched interfaces.\(^{35}\) Pathway [E] had been proposed for explaining the underestimation of thermal conductance by DMM for a Pb-Diamond interface,\(^{37}\) however, it was shown later that this could not be the possible mechanism.\(^{35}\) Pathway [D] has been shown to be a probable cause for lowering of experimental conductance as compared to DMM.\(^{36}\) According to this theory, there is a mixing zone near the interface where the properties are intermediate to the two substrate materials. It has been shown that additional scattering of phonons in this zone lowers the probability of transmission and reduces the thermal conductance.
In this work, we will consider the pathway [A]+[B]. Thermal conductance for pathway [B] is given by DMM as discussed previously. Now we will focus our attention on calculating electron-phonon thermal conductance for pathway [A] which is referred to as $G_{e-ph}$ here. The commonly cited approach to calculate $G_{e-ph}$ is to consider a two-temperature model\cite{19,20,21} where electrons at temperature $T_e$ exchange energy with phonons at temperature $T$. Under conditions of strong non-equilibrium between the two, the energy transfer rate becomes proportional to the difference of the fifth power of the temperatures\cite{20}. Assuming a small departure from equilibrium, an energy
balance based on the Fourier law\(^7\) yields an electron-phonon thermal conductance, \(G_{e-ph} = \sqrt{G k_p}\), where \(G\) is the electron cooling rate and \(k_p\) is the phonon thermal conductivity of the metal. As explained below, the latter model remains largely empirical since \(G_{e-ph}\) is commonly fit to the interfacial thermal conductance data. In this work, we adopt an alternate approach to obtain \(G_{e-ph}\) directly from the Bloch-Boltzmann-Peierls formula for electron-phonon interaction that provides an improved assessment of the role of the electron-phonon thermal conductance in interfacial thermal transport.

As discussed above, a commonly used approach in the literature is to express the electron-phonon thermal conductance as \(G_{e-ph} = \sqrt{G k_p}\). However, the phonon thermal conductivity of the metal, \(k_p\), is usually unknown due to the dominance of the electronic contribution to thermal conductivity. To proceed with this model, the electron-phonon conductance is assumed to be of the form \(G_{e-ph} = \psi/\sqrt{T}\), where \(\psi\) is an empirical constant obtained from fitting thermal conductance data. This approach yields the value of \(G_{e-ph}\) within a range of 0.3-1 GW/m\(^2\)K. Overall, the model is ambiguous since the empirical parameter needed to calculate electron-phonon thermal conductance must be extracted from the total interfacial conductance after accounting for the phonon-phonon conductance estimated from DMM. Further, the assumed temperature dependence \(k_p \sim T^{-1}\) used in such calculations is only valid for temperatures above the Debye temperature. A more accurate description would require the fitting of the parameter using the temperature dependence of \(k_p\) in three different regimes: \(T \ll \theta_D, T \gg \theta_D\) and \(T \sim \theta_D\) where \(\theta_D\) is the Debye temperature of the metal.

Here, we introduce an approach to obtain \(G_{e-ph}\) valid over a wide temperature range without requiring any fitting to the thermal conductance data. Our method uses an empirical deformation
potential to describe the matrix element, but the approach remains advantageous in that the empirical fit is to electrical resistivity data and is decoupled from the thermal conductance data. Thus, the predicted electron-phonon thermal conductance is independent of the validity/reliability of the DMM predictions of the phonon-phonon thermal conductance. We argue that this provides a better answer to the question whether electron-phonon coupling can indeed explain the discrepancy between the experimental thermal conductance of metal-dielectric interfaces and predictions of the theoretical diffuse mismatch model (DMM).

We start our formulation by assuming that electrons are in equilibrium at temperature $T_e$ and the phonons are in equilibrium at temperature $T$. Electrons from an initial quantum state defined by wave vector $k$ move into final quantum state $k'$ and release/absorb a phonon in the process. Momentum and energy conservation for the system requires:

$$k' = k \pm q$$

$$\frac{\hbar^2 k'^2}{2m^*} = \frac{\hbar^2 k^2}{2m} \pm \hbar \omega_q$$

(3.6)

where $q$ and $\omega_q$ are the phonon wave vector and frequency respectively. The transition probability for an electron to go from initial state $|k\rangle$ to a final state $|k'\rangle$ is calculated from Fermi’s golden rule and is given by:

$$P(k, k') = \frac{2\pi}{\hbar} \left| \langle k' | H | k \rangle \right|^2 \delta \left( E_{k'} - E_k \right)$$

(3.7)

Where $E_{k'}$ and $E_k$ are the energies of the final and initial states and the $\delta$-function ensures energy conservation. Using the above transition probability, conservation laws and Bloch-Boltzmann-Peierls formula we get the net rate of change of the phonon occupation due to electron scattering:
\[ \frac{\partial n_q}{\partial t} = \frac{2\pi}{\hbar} \sum_k \left[ \langle k | H | k + q \rangle^2 \delta \left( E_{k,n_q+1} - E_{k+q,n_q} \right) f(k+q) \left( 1 - f(k) \right) - \langle k + q | H | k \rangle^2 \delta \left( E_{k+q,n_q-1} - E_{k,n_q} \right) f(k) \left( 1 - f(k+q) \right) \right] \]  

(3.8)

where \( n_q \) is the Bose-Einstein occupation number for the phonons with wave vector \( q \), \( k \) is the initial electron wave vector, \( H \) is the interaction Hamiltonian for electron-phonon scattering, \( f \) is the Fermi distribution given by 

\[ f(k) = \frac{1}{1 + e^{-E(k)/k_B T}} ; E(k) = \frac{\hbar^2 k^2}{2m^*} \]  

and \( E \) is the energy of the electron-phonon system.

We employ deformation potential theory\(^{42,43}\) to describe the interaction Hamiltonian for electron-phonon scattering process as

\[ H = D_{ac} \frac{\delta V}{V} \]  

(3.9)

where \( D_{ac} \) is the deformation potential for electron scattering by acoustic phonons, \( V \) is the volume and \( u(r) \) is the displacement vector at position \( r \). Using the Hamiltonian of Eqn. (3.9) the matrix element for electron-phonon interaction can be written as:

\[ \left| \langle k | H | k \rangle \right|^2 = \frac{D_{ac}^2 \hbar Q_q}{2Vd_v^2} \left( n_q + \frac{1}{2} \pm \frac{1}{2} \right) = \frac{D_{ac}^2 \hbar q}{2Vd_v} \left( n_q + \frac{1}{2} \pm \frac{1}{2} \right) \]  

(3.10)

where \( d \) is the density of the metal and \( v_s \) is the speed of longitudinal waves. Here we use the Debye density of states for phonons. The positive sign inside the brackets indicates phonon emission and the negative sign indicates phonon absorption.

The electron-phonon scattering time can be expressed in terms of the deformation potential as\(^{43}\)
Using the known relation between the electrical resistivity due to phonon scattering and the average electron scattering time: 
\[ \rho_{e-ph} = \frac{m^*}{n e^2 \tau(E_F)} \]
we get a direct relation between resistivity and the deformation potential. This relation is,

\[ D_{ac} = \frac{2\pi \hbar^4 \nu_e^2 n e^2}{(2\pi)^{\frac{3}{2}}(m^*)^\frac{5}{2}k_B^2} \left( \frac{\rho_{e-ph}}{T} \right) \left( \frac{1}{\sqrt{E_F}} \right) \]

where \( \rho_{e-ph} \) is electrical resistivity omitting the low temperature residual resistivity and can be readily obtained from the total resistivity using Matthiessen’s rule. We have calculated the best-fit value for \( D_{ac} \) for six metals (Au, Al, Ag, Cr, Cu and Pt) using multiple linear regression over the temperature range and found the values to be 4.50, 6.37, 3.70, 14.70, 5.22 and 10.12 eV respectively. For \( T > \theta_D \), the variation in \( D_{ac} \) with temperature is <10\% for these metals. The discrepancy between \( G_{e-ph} \) calculated using the exact temperature dependent \( D_{ac} \) compared to that from the best fit temperature independent value is also <10\% for all the metals listed here except for Au and Cr where it is as much as 40\%. The temperature independence can be understood by considering, for example, the Bloch-Gruneisen formula for electrical resistivity due to electron-phonon scattering:

\[ \rho_{e-ph} = A \left( \frac{T}{\theta_D} \right)^5 \frac{x^5}{(e^x - 1)(1-e^{-x})} dx \]

This yields \( D_{ac} \) independent of \( T \) for \( T > \theta_D \), with

\[ D_{ac}^2 = \frac{2\pi \hbar^4 \nu_e^2 n e^2}{2^\frac{3}{2} \sqrt{m^* k_B}} \frac{1}{\sqrt{E_F}} \frac{A}{4\theta_D} \].

In our calculations, we found
$D_{ac}$ to be essentially constant for $T > 0.1\theta_D$. When $T \ll \theta_D$, 

$$D_{ac}^2 = \frac{2\pi \hbar^4 \nu^2 e^2}{2^3 m^* \mu_b^2} \frac{1}{\sqrt{E_F}} A T^4 \theta_D^{1/T} \int_0^{\infty} \frac{x^5}{(e^x - 1)(1 - e^{-x})} dx \text{ and is temperature dependent.}$$

To simplify Eqn. (3.3) we covert summation over $k$ space into integral as:

$$\sum_k F(k) = \int_{k=(q/2)}^{\infty} \int_{k=(q/m^*_\nu)}^{\pi} \int_{\theta=0\phi=0}^{2\pi} \frac{2V}{(2\pi)^3} F(k) k^2 \sin \theta \, d\theta \, d\phi \, dk \quad (3.13)$$

The lower limit on $k$ comes from energy conservation. The additional factor of 2 along with the usual factor of $V/(2\pi)^3$ comes from the fact that electrons of both spins can participate in scattering. After the integration over space, Eqn. (3.8) simplifies to:

$$\frac{\partial n_q}{\partial t} = \frac{D_{ac}^2 m^*}{4\pi^2 \hbar^2 \nu} \int_{q=(q/2)}^{\pi} \int_{k=(q/m^*_\nu)}^{\infty} dk \, k \left[ \delta_q + f(k+q)(1-f(k)) \right]$$

$$n_q f(k)(1-f(k+q))] \quad (3.14)$$

At room temperature, the term $(m^*_\nu/\hbar)$ is small compared to $(q/2)$ and can be neglected. Finally, the net rate of energy exchange between electrons and phonons is

$$\frac{dQ}{dt} = \sum_q \hbar \nu_q \left( \frac{\partial n_q}{\partial t} \right) \quad (3.15)$$

$$= \left( \frac{\hbar \nu V}{2\pi^2} \right) \int_{q=0}^{\infty} q^3 \left( \frac{\partial n_q}{\partial t} \right) dq$$

where $q_D$ is the Debye wave vector. Expressing the energy balance between electrons and phonons as

$$\frac{dQ}{dt} = G_{e-ph} A(T_e - T) \quad (3.16)$$
where $A$ is the cross-sectional area. We then obtain the electron-phonon conductance as

$$G_{e-ph} = \frac{\hbar V}{2\pi^2 A(T_e - T)} \int_{q_0} q^3 \left( \frac{\partial n_q}{\partial t} \right) dq = \frac{\hbar L V}{2\pi^2 (T_e - T)} \int_{q_0} q^3 \left( \frac{\partial n_q}{\partial t} \right) dq \quad (3.17)$$

where $L$ is the characteristic length scale for energy transfer. Here we choose $L = f \Lambda_e$ where $\Lambda_e$ is the electron mean free path in the metal. We use a value of 3 for $f$ corresponding to ~95% energy loss. We note that this is different from the effective length scale for energy transfer in Ref. [10] where it is given by $l \approx \left( \frac{k_p}{G} \right)^{1/2}$. In the latter case, the length scale is only a fraction of the electron mean free path and seems unphysical. Eqn. (3.12) can be simplified using Eqn. (3.17) as

$$G_{e-ph} = \frac{D_{ac}^2 m^2 k_B T_e L}{4\pi^2 \hbar^3 d (T_e - T)} \int_{q_0} q^3 \left( \frac{\hbar v_q}{e^{k_BT_e} - 1} - \left( \frac{\hbar v_q}{e^{k_BT_e} - 1} \right)^{-1} \right) \left\{ \ln \left( 1 + e^{k_BT_e} \frac{E_F - \frac{k_B T_e}{m^*}}{e^{k_BT_e} - \frac{k_B T_e}{m^*}} \right) \right\} dq \quad (3.18)$$

The above expression can be further simplified by expanding the first term inside the integral in the limit of high temperatures.

In subsequent numerical calculations, we employ the Fermi-Dirac and Bose-Einstein distribution functions to obtain $G_{e-ph}$ across a wide temperature range. We have verified that the computed value of $G_{e-ph}$ at 300 K is independent of the temperature difference $(T_e - T)$. Figure 3.2 shows the variation of $G_{e-ph}$ with temperature for the six metals up to 500 K. The value of $G_{e-ph}$ increases with temperature and becomes approximately constant at room temperature. The values range between ~0.1-4 GW/m$^2$K at room temperature. Specifically, the values at room temperature for Au and Al are 0.11 GW/m$^2$K and 1.1 GW/m$^2$K respectively. We compare these
with values based on the model of Ref. [10] using measured values of $k_p^{45,46}$ and electron cooling rates from the two-temperature model.\textsuperscript{47,48} The latter values are 0.29 GW/m$^2$K and 2.21 GW/m$^2$K respectively\textsuperscript{49} and are of the same order of magnitude as our predictions but approximately a factor of 2 larger. We have performed the calculations across a wide temperature range, both $T<\theta_D$ and $T\sim\theta_D$. For $T<\theta_D$, our calculations show that $G_{e-ph}\sim T^n$, where the exponent $n$ ranges between 3.5-3.7 expect for Au where the exponent is 3.1. This is in agreement with the predicted dependence of $G_{e-ph}$ from the two-temperature model\textsuperscript{38} where the dependence is $G_{e-ph} = \sqrt{Gk_p} \sim \sqrt{T^4T^3} \sim T^{3.5}$. We do not compare the temperature trends for $T\sim\theta_D$ since $G$ does not have a simple analytical power law dependence on $T$ for $T\sim\theta_D$.\textsuperscript{50}

![Figure 3.2. Thermal conductance due to electron-phonon coupling, $G_{e-ph}$ as a function of temperature for six metals. The temperature dependence of $G_{e-ph}$ closely follows $\sim T^{3.5}$ at low temperatures ($T/\theta_D<0.1$).](image-url)
We now investigate the relative contribution of the electron-phonon conductance to the net thermal conductance for several metal-dielectric interfaces. The diffuse mismatch model provides $G_{ph-ph}$, the thermal conductance due to the energy transfer between phonons. The net conductance with $G_{e-ph}$ and $G_{ph-ph}$ in series is

$$G_{net} = \frac{G_{e-ph}G_{ph-ph}}{G_{e-ph} + G_{ph-ph}}$$

(3.19)

We have computed $G_{net}$ as a function of temperature for all the six metals mentioned above with silicon, silica, sapphire and diamond as the dielectric. For interfaces whose vibrational spectra are similar (Debye temperatures approximately equal), $G_{DMM}$ is considered to be a reasonable estimate of the phonon-phonon interfacial thermal conductance. In such cases, the discrepancy between $G_{DMM}$ and $G_{net}$ from measurements varies between ~3-7 times at room temperature.\(^6\)\(^5\)\(^1\) In our calculations, $G_{DMM}$ never exceeded $G_{net}$ by a factor greater than 2 for $T>200$ K. The maximum remains 1.8 for an Al-SiO\(_2\) interface. Figure 3.3 plots the absolute difference between $G_{net}$ and $G_{DMM}$ as a fraction of $G_{DMM}$ for silicon and sapphire interfaces with Al, Au, Cr and Pt. Thus, our numerical calculation of electron-phonon thermal conductance shows that the electron-phonon coupling is not a plausible reason for the observed discrepancy at room temperature. Our theory also extends to $T<\theta_D$ where $G_{DMM}$ exceeds the calculated total thermal conductance by a factor of up to 5 but this factor falls off quickly to less than 2 as we reach 50 K. Thus, our results are consistent with recent work that suggests other factors such as interface defects\(^5\)\(^6\)\(^36\) are responsible for the reduction in $G_{net}$. 
We note that while the analysis in this work assumes linear phonon dispersion, it is fairly straightforward to extend the analysis to non-linear dispersion. We have verified that this does not alter the conclusion and $G_{e-ph}$ predictions remain of the same order of magnitude. We considered a more accurate sinusoidal dispersion. The comparison between linear and sine dispersion for Au is shown in figure 3.4. The values of $G_{e-ph}$ increase from those corresponding to linear dispersion by as much as 180% for Al and <140% for other metals. For $T>200$K, the increase is modest, between 25-65% across all the metals considered in our work. Previous work\textsuperscript{5} has considered the effect of non-linear phonon dispersion on $G_{DMM}$ for Au and Al interfaces with diamond and sapphire, finding that values decrease by a factor as much as 3.7. Consequently, the inclusion of non-linear dispersion would increase the ratio of $G_{e-ph}$ to $G_{DMM}$ and render the electron-phonon thermal conductance even more insignificant. Our conclusions thus remain the same irrespective of phonon dispersion. Thus by studying the net thermal conductance across several metal-dielectric interfaces, we find that electron-phonon coupling does not add significant thermal resistance to the DMM resistance at temperatures above 200 K.
We hypothesize that the discrepancy between the experimental and DMM values of thermal conductance is likely due to other factors such as interface substrate damage, interfacial mixing and interfacial defects due to lattice mismatch. We conclude that electron-phonon coupling for such interfaces can be ignored in considerations of thermal transport.

Figure 3.4. Comparison between $G_{e-ph}$ calculated using linear and more accurate sine dispersion for gold.

Even though we have shown that electron-phonon thermal conductance can be ignored due to its higher value as compared to DMM value, we include it in the subsequent analysis for better accuracy. We have calculated $G_{net}$, the harmonic sum of $G_{e-ph}$ and $G_{DMM}$. To calculate $G_{microscopic}$, we scale $G_{net}$ with the contact area ratio as:
\[ G_{\text{microscopic}} = G_{\text{net}} \times \pi a^2 \eta \int_d^\infty \phi(z) dz \] (3.20)
4. NET THERMAL CONDUCTANCE: THEORETICAL v/s OBSERVED

4.1 Net thermal conductance for transfer printed interfaces

We have calculated $G_{\text{macroscopic}}$ and $G_{\text{microscopic}}$ in the previous chapters. These two conductances act in series and as a result, the net thermal conductance for a transfer printed interface is given by:

$$G = \frac{G_{\text{macroscopic}} \times G_{\text{microscopic}}}{G_{\text{macroscopic}} + G_{\text{microscopic}}} \quad (4.1)$$

We now compare the results of the above theory with experimental data\textsuperscript{12} on interfacial thermal conductance of Au-SiO\textsubscript{2} and Au-Si interfaces in the temperature range 300-600 K. Figure 4.1 plots the experimental and theoretical conductance across the two interfaces. Wet and dry adhesion result in different areas of contact and hence, differing thermal conductances. Further, the inclusion of geometric scattering in the phonon-phonon conductance provides a lower limit to conductance as discussed previously and is plotted separately in the case of Au-SiO\textsubscript{2}. Inclusion of ballistic effects reduces the conductance by a third. However, this will only be present at certain locations and provides the lower bound on thermal conductance. Overall, wet adhesion augments the thermal conductance significantly, more than doubling the conductance at 300 K, in comparison to dry adhesion.
Figure 4.1 Comparison of theory with experimental data on the interfacial thermal conductance between (a) Au-SiO$_2$ and (b) Au-Si as a function of temperature. Wet adhesion significantly enhances conductance but the predictions still underestimate the measured values.
The predicted conductance for wet adhesion is of the same order of magnitude as the measured value indicating the significance of plastic deformation in providing a relatively large area of contact for heat conduction. The predicted value for Au-SiO₂ varies between ~18 MW/m²K at 300 K to ~9 MW/m²K at 600 K. The decreasing temperature trend in the case of wet adhesion agrees well with the general trend of the experimental data giving further credence to the argument that wet adhesion is involved in these interfaces. Temperature dependence arises entirely due to temperature dependent mechanical properties that affect deformation mechanics. Standard DMM predicts an approximately constant conductance at temperatures above the Debye temperature. In the case of Au-Si, the predictions again underestimate the data. The somewhat larger difference is due more to the smaller DMM predictions rather than a dramatically reduced role of plastic deformation. Overall, there remains a significant quantitative discrepancy between theory and experiments in both these interfaces.

4.2 Conduction through capillary bridges

We speculate that the discrepancy arises due to heat conduction via capillary bridges. Previous ultrafast thermoreflectance experiments on the Au-water interface measured \( G > 20 \) MW/m²K. The conductance increases as the surface becomes more hydrophilic. While low frequency phonons transport heat independent of the interfacial bonding, high frequency phonons transmit better as the interface bonding strengthens. Molecular simulations also corroborate the measurements and further reveal that the conductance is directly proportional to the work of adhesion. An upper limit to the conductance is unknown at present but the value can exceed 100 MW/m²K.
Assuming that water capillaries conduct heat parallel to solid-solid contacts, the product of the net conductance of capillary bridges and the fractional area available for such conduction, \( G_{\text{capillary}} \times A_{\text{capillary}} / A_n \), should equal \( \sim 30 \text{ MW/m}^2\text{K} \) in the case of Au-SiO\(_2\) at 300 K. The latter is the net difference in the measured conductance and the calculated conductance in Fig. 4.1(a). Assuming a similar areal fraction under water capillary as the solid-solid contact, the required conductance across the capillary is \( \sim 120 \text{ MW/m}^2\text{K} \). This appears reasonable based on existing experiments\(^{49,53}\) and simulations\(^{54}\) where the reported range is 20-300 MW/m\(^2\)K. The actual areal coverage of water capillaries is difficult to estimate directly\(^{55}\) based on the current state of research, especially for a distribution of asperities across different length scales. Since the average separation between surfaces is <1 nm (comparable to the roughness deviation), the behavior of water is likely not that of the bulk. While theories for the structure of water at sub-nm asperities exist\(^{56}\), considerations of heat conduction through such structures are absent in the literature. More insight in this problem requires measurements with controlled humidity in tandem with molecular simulations. We do note that existing molecular simulations of a broad range of surface chemistries suggest the absence of a water vapor layer at the interface even when hydrophobic\(^{54,56-58}\), suggesting that conduction via water capillaries could contribute significantly.

Finally, we present the estimated thermal conductance for different metal-dielectric interfaces assuming similar roughness profiles. Figure 4.2(a) shows the conductance for different metals with SiO\(_2\) as the substrate. The figure shows both wet and dry adhesion. The values are on the order of \( 10 \text{ MW/m}^2\text{K} \) for the case of wet adhesion, suggesting good conductance for most metals used in transfer printing. The variation amongst metals is largely the consequence of
differing plasticity indices. Figure 4.2(b) plots the conductance between Au and various substrates. The variation in this case is related more to phonon-phonon conductance.
Figure 4.2 Interfacial thermal conductance at room temperature for various metal-dielectric interfaces. (a) The calculations are for different metals with SiO2 as the substrate at room temperature. Al shows the highest interfacial thermal conductance and Au the lowest. (b) The same calculation for the interface between Au and various substrates.
5. SUMMARY AND CONCLUSIONS

In this work, we have considered two of the main factors controlling the interfacial thermal conductance for metal-dielectric interfaces: electron-phonon coupling in the metal and fractional area coverage at the interface. The effect of electron-phonon coupling on the thermal conductance for metal-dielectric interfaces is not clearly understood. The leading model is semi-empirical in nature and it depends on the thermal conductance measurements for the calculation of empirical parameter. We have developed a model of the thermal conductance due to electron-phonon coupling that can be calculated independent of thermal conductance measurements. The current work is based on the scattering theory of electrons and phonons and the empirical parameter needed for calculation is extracted from the electrical resistivity data that is more reliable than the procedure adopted in the previous model. We find $G_{e-ph}$ to range between 0.1-4 GW/m²K at room temperature across six different metals. For $T<\theta_D$, the temperature dependence of $G_{e-ph}$ predicted by our model is in agreement with that from the two-temperature model. By studying the net thermal conductance across several metal-dielectric interfaces, we find that the electron-phonon coupling does not add significant thermal resistance to the DMM resistance at temperatures above 200 K. We hypothesize that the discrepancy between the experimental and the DMM values of the thermal conductance is then likely due to other factors such as interface substrate damage, interfacial mixing and interfacial defects due to lattice mismatch. We conclude that electron-phonon coupling for such interfaces can be ignored in considerations of thermal transport and hence, provide an answer to whether the electron-phonon thermal conductance should be considered for metal-dielectric interfaces. Based on our work, the microscopic thermal conductance can be directly estimated by the DMM.
Macroscopic component which is due to the constriction of actual contact area as compared to the nominal area, gains relative importance in limiting thermal transport for rough interfaces. For transfer-printed Au-dielectric interfaces with sub-nanometer roughness, we have shown that the plastic deformation of Au, in combination with capillary forces leads to a large fractional area of coverage (~0.25). Such a high value of fractional area coverage shows that the transfer printed interfaces are not at a big disadvantage as compared to the interfaces formed by vacuum deposition in terms of contact area. From the comparison of fractional area coverage for dry and wet adhesion, we conclude that the capillary forces augment the contact area by more than two times as compared to dry adhesion at room temperature. Also, the temperature trends of contact area for wet and dry adhesion cases are different and opposite in nature.

Finally, the net thermal conductance has been calculated as the series sum of macroscopic and microscopic thermal conductances. The calculations show the value of interfacial conductance to be ~10 MW/m²K, comparable in magnitude to the experimental data. The temperature trend of net thermal conductance further agrees with experimental data giving credence to wet adhesion hypothesis. We attribute the discrepancy between theory and data to conduction via water capillary bridges. We support this hypothesis with a preliminary numerical analysis. Further experimental and theoretical verification remains a topic of future research.
REFERENCES


