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Effect of interface adhesion and impurity mass on phonon transport at atomic junctions

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With the characteristic lengths of electronic and thermal devices approaching the mean free paths of the pertinent energy carriers, thermal transport across these devices must be characterized and understood, especially across interfaces. Thermal interface conductance can be strongly affected by the strength of the bond between the solids comprising the interface and the presence of an impurity mass between them. In this work, we investigate the effects of impurity masses and mechanical adhesion at molecular junctions on phonon transmission via non-equilibrium Green's functions (NEGF) formalisms. Using NEGF, we derived closed form solutions to the phonon transmission across an interface with an impurity mass and variable bonding. We find that the interface spring constant that yields the maximum transmission for all frequencies is the harmonic mean of the spring constants on either side of the interface, while for a mass impurity, the arithmetic average of the masses on either side of the interface yields the maximum transmission. However, the maximum transmission for each case is not equal. For the interface mass case, the maximum transmission is the transmission predicted by a frequency dependent form of the acoustic mismatch model, which we will refer to as the phonon mismatch model (PMM), which is valid for specular phonon scattering outside the continuum limit. However, in the interface spring case, the maximum transmission can be higher or lower than the transmission predicted by the PMM. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4773331]

I. INTRODUCTION

The ability to accurately predict thermal conductance is essential for effective design of nano-structured devices. Interfaces can dominate thermal resistance and severely limit thermal transport as these devices decrease in size and approach length scales comparable to the mean free paths of pertinent thermal energy carriers. The two most widely used models to predict thermal conductance across interfaces are the acoustic mismatch model (AMM) and the diffuse mismatch model (DMM), both of which are based solely on the bulk properties of the materials on either side of the interface. However, the effect of these interfaces is not only dependent on the properties of the materials on either side of the interface, but also the interface properties, including chemistry,[1–3] mixing,[4,5] dislocations,[3,6] and bonding[7–15] at the interface.

Many studies have also shown that careful manipulation of the interface properties can be used to tune the thermal boundary conductance.[1,4–6,16–19] For example Hopkins *et al*.[4] measured the thermal boundary conductance (TBC) of several Cr/Si interfaces with varied amounts of interfacial mixing and showed that the effects of interface mixing can dramatically affect the thermal transport across the interface. Hopkins et al. also showed that oxide layers on Si (Ref. 1) and dislocations at Al/GaSb and GaSb/GaAs interfaces[6] play a major role in TBC leading to decreases in TBC of 50 MW m^{-2} K^{-1} and 10 MW m^{-2} K^{-1}, respectively. Collins et al.[2] showed that terminating chemistry on the surface of synthetic diamond interfaces impacts interface thermal transport. Hopkins et al.[8] showed that by chemically functionalizing graphene surfaces with adsorbates, they were able to vary thermal conductivity by a factor of two which was attributed to a change in the density of covalent bonding. Recently, Losego et al.[19] showed that changing the terminating group on self-assembled monolayers affected thermal conductance by a factor of nearly two by doubling the bond strength.

At the same time, several studies have investigated the effect of interface properties on TBC through numerical and theoretical methods.[7,9,11,12,14,15] For example, Shen et al.[11] showed that it is possible to augment thermal conductance by applying pressure to coupled systems of dissimilar materials. For the weakly coupled systems, they were able to increase the conductance with increasing pressure by an order of magnitude until they reached a plateau in the conductance. Ong and Pop[14] investigated the effect on van der Waals interaction strength on conductance between carbon nano-tubes and found that the conductance varied linearly with interaction strength. Zhang et al.[15] studied the effect of

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bonding on phonon transmission and conductance one-dimensional harmonic junctions between dissimilar materials. They found that a peak in transmission and thus a peak in conductance occurs when the harmonic bond strength between the two materials was equal to the harmonic mean of the bonds strengths within the materials on either side of the interface. The impact of the mass and bond imperfection at interfaces is clearly an important consideration in the fundamental physics of TBC. Understanding these effects could allow device engineers to predict and tune TBC through the manipulation of bonding at an interface and the introduction of thermal interface materials.

A large subset of the previously mentioned studies focused on bonding and demonstrated that stronger bonding resulting in greater conductance. However, the results of these studies were a convolution of many effects such as changes in bond strength, mass, order, and lattice parameter. For example, when Hopkins et al.[8] changed the bonding between graphene and Al via chemical functionalization, they not only changed the bond strength, they added impurity masses. Along the same lines, when Losego et al.[19] augmented the effective SAM-Au adhesion strength through a change in the terminating group of the SAM, he changed both the bonding and mass of the terminating group. This convolution of effects makes it difficult to determine how each properties contributes to TBC and how to choose each property to optimize the interface of interest.

Therefore, in this study, we separately investigate the effects of bonding and impurity masses at molecular junctions on phonon transmission using a non-equilibrium Green's functions (NEGF) formalism and derive analytical solutions for phonon transmission across one-dimensional atomic interfaces. We find that for the single impurity mass case, setting the mass equal to the arithmetic mean of the masses on either side of the interface yields the greatest transmission for all frequencies. When considering interface bonding, we find that setting the interface spring constant to the harmonic mean of the springs on either side of the interface yields the greatest transmission for all frequencies. However, we find that coupling the interface with a spring and coupling with a mass are not always equal. For instance, the maximum transmission that can be achieved when coupling with a mass is the transmission predicted by a frequency dependent form of the AMM which we term the phonon mismatch model (PMM) for reasons that will be clarified in Sec. III. However, when coupling with a spring, the maximum transmission can be greater or lower than the PMM transmission depending on the properties of the materials comprising the interface.

II. NON-EQUILIBRIUM GREEN'S FUNCTION FORMALISM

The formulation for NEGF (as it is applied here) is based on a system of classical harmonic Newtonian equations which describes the forces acting on each atom in a one-dimensional model system. The model system is comprised of three parts: a semi-infinite source and sink, and a center region. If we assume that the solutions to the system of harmonic Newtonian equations will be in the form of plane waves, the displacement of the nth atom can be described by

$$u_n = B \exp[iqna] + C \exp[-iqna], \tag{1}$$

where q is the wavevector, a is the material-dependent interatomic spacing, and B and C are the unknown amplitudes of the forward and backward traveling waves, respectively. In general, the wavevector is dependent on frequency, so in order to solve for the frequency-dependent transmission coefficient, we must assume a form of the dispersion. Since we are dealing with a 1D system of springs and masses and a single atom basis, the dispersion is sine type.[20,21] Representing the system of equations in matrix form and solving for the vector of displacements yields, $u = SG$, where u is the column vector of displacements, S is the source column vector, and G is the Green's function matrix. The Green's function matrix is

$$G = [M\omega^2 - K - \Sigma_1 - \Sigma_2]^{-1}, \tag{2}$$

where M is the diagonal mass matrix, K is the tridiagonal spring constant matrix, and Σ_j is the self energy matrix for the terminal atom on the jth side of the channel, i.e., side 1 or side 2. The self energy matrix, Σ_j, can be rewritten as

$$\Sigma_j = -k_j \exp\left(2i \sin^{-1}\left(\frac{\omega}{\omega_{c,j}}\right)\right), \tag{3}$$

where ω_c is the cutoff frequency. The elements $K_{p,q}$ in the spring constant matrix represent the spring constants between the pth atom and qth atom and in general, $K_{p,q} = K_{q,p}$ or $K = K^T$. The frequency broadening matrix is given by

$$\Gamma_j = i(\Sigma_j - \Sigma_j^\dagger). \tag{4}$$

Using Eq. (3), the non-zero element of Eq. (4) can be rewritten as

$$\Gamma_j = 2\omega\sqrt{k_j m_j - \frac{m_j^2 \omega^2}{4}}. \tag{5}$$

In crystals, the acoustic impedance in a specific crystallographic direction, $Z(0)$, is defined as the group velocity at the Brillouin zone center, $v_g(0)$, times the crystal mass density, ρ. Defining the phonon impedance as the "acoustic" impedance extended outside of the long wavelength limit, the phonon impedance is $Z(\omega) = \rho v_g(\omega)$. In one-dimension with a sine-type dispersion, $\rho = m/a$ and $v_g = a\sqrt{\frac{k}{m}}\cos\left(\frac{qa}{a}\right)$ so that the phonon impedance becomes

$$Z_j = \sqrt{k_j m_j - \frac{\omega^2 m_j^2}{4}}. \tag{6}$$

Therefore, Eq. (5) is the frequency dependent phonon impedance of the one-dimensional semi-infinite side j multiplied by 2ω. It can be shown that this goes to zero as we approach the Brillouin zone edge or as we approach the cutoff frequency due to the choice of a sinusoidal dispersion. In addition at low frequencies, $\omega \to 0$, Eq. (6) reduces to $Z(0) = \sqrt{k_j m_j}$, the acoustic impedance.

The NEGF frequency dependent transmission coefficient is defined by the Caroli formula,[22]

$$T(\omega) = Trace[\Gamma_1 G \Gamma_2 G^\dagger], \quad (7)$$

where $G^\dagger$ is the adjoint of the Green's function matrix. A detailed description of the NEGF formalism for modeling phonon transport is described in Refs. 23 and 24. We have only presented a brief overview of the method here.

III. ONE ATOM CHANNEL TRANSMISSION

In a one-dimensional system, there are only two ways of coupling two control volumes using a single element; either with an interface mass or an interface spring. In this section, we investigate the first case. Figure 1 shows the model used to calculate the transmission across a junction with an impurity mass. In this case, we only consider one mass in our NEGF derivation, which is in general different than the masses in the source and sink. Note, that k_1 is the same spring as in the source and k_2 is the same spring that is in the sink, and in general k_1 and k_2 are not the same. Following the steps outlined in Sec. II, we find that the phonon transmission across an interface coupled by an interface mass is

$$T(\omega, m_{\text{int}}) = \frac{4\Gamma_1\Gamma_2}{(\Gamma_1 + \Gamma_2)^2 + 4\omega^4\left(\frac{m_1+m_2}{2} - m_{\text{int}}\right)^2}. \quad (8)$$

We begin our investigations of this transmission numerically with Fig. 2(a), which plots transmission versus frequency for several values of m_{int}. Mass, spring constant, and frequency are normalized by 1.67×10^{-27} kg, 1.67×10^{-3} N/m, and THz, respectively. As can be seen in Fig. 2(a), as m_{int} increases, the transmission for all frequencies increases until m_{int} reaches a certain value, at which point it decreases until it reaches zero. This suggests that there exists an interface mass that maximizes transmission for all frequencies. In order to investigate this effect further, we plot transmission versus m_{int} for several frequencies in Fig. 2(b). It becomes clear that the transmission is maximized at the same value of m_{int} for all frequencies. We can also see that lower frequency phonons are affected less by a change in the interface mass than higher frequencies.

Taking the limit of Eq. (8) as $\omega \to 0$, we find that the transmission becomes a constant which is independent of m_{int},

$$T_{AMM} = \frac{4\sqrt{k_1 m_1 k_2 m_2}}{(\sqrt{k_1 m_1} + \sqrt{k_2 m_2})^2} = \frac{4Z_1(0)Z_2(0)}{(Z_1(0) + Z_2(0))^2}, \quad (9)$$

where $Z(0) = \sqrt{km}$ is the acoustic impedance in one-dimension. Equation (9), then, is the transmission predicted by
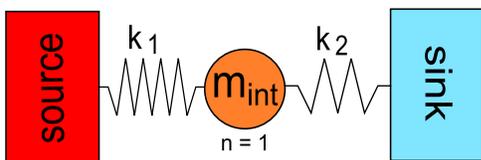

FIG. 1. Model system used to calculate the transmission across the interface between two dissimilar crystalline materials with an impurity mass at the interface. Mass, spring constant, and frequency are normalized by $1.67\,10^{-27}$ kg, $1.67\,10^{-3}$ N/m, and THz, respectively.
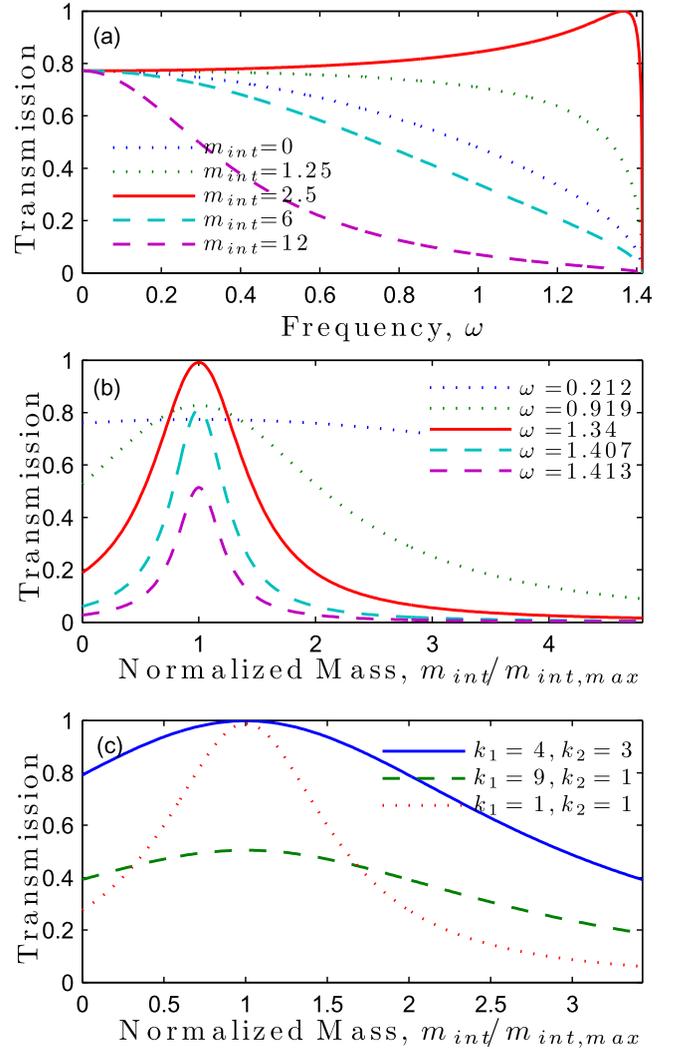

FIG. 2. Predicted transmission calculated using Eq. (8) using $k_1 = 1$, $k_2 = 2$, $m_1 = 1$, $m_2 = 4$, and $m_{\text{int,max}} = 2.5$. Mass, spring constant, and frequency are normalized by $1.67\,10^{-27}$ kg, $1.67\,10^{-3}$ N/m, and THz, respectively. (a) Plotted versus frequency for various values of m_{int}. The dashed lines are for values of m_{int} greater than 2.5 and dotted lines for values lower than 2.5. (b) Plotted versus m_{int} for various frequencies. A maximum transmission occurs for all frequencies when $m_{\text{int}} = 2.5$. (c) Transmission versus normalized mass for $\omega = 0.9$ and various choices of k_1 and k_2. The mass which yields maximum transmission is independent of the spring constants.

the AMM in the one-dimensional long wavelength case. Note that low frequency phonons are affected by the interface mass as the interface mass approaches infinity since $m_{\text{int}} = \infty$ creates an immovable barrier placed between the two control volumes.

We also observe that the transmission is symmetric on either side of $m_{\text{int,max}}$ for all frequencies. This means that negative and positive perturbations from the ideal interface mass will have the same effect on the transmission. The same result was found for elastic scattering with an impurity mass in a periodic crystal field by Ref. 25, one of Klemens' seminal works on the elastic scattering of phonons by static imperfections. Since Ref. 25 and our formalism only consider elastic scattering, this result is consistent. From inspection of Eq. (8), transmission is a symmetric function in m_{int}.

We were able to further validate this result by finding the analytical maximum of Eq. (8) with respect to m_{int}, so that the mass which maximizes transmission is

$$m_{\text{int,max}} = \frac{m_1 + m_2}{2}. \quad (10)$$

This shows that the interface mass which yields the maximum transmission for all frequencies is the arithmetic mean of the two masses on either side of the interface. We also notice that this mass is independent of the spring constants on either side of the interface even though the transmission across the interface, Eq. (8), is dependent on these spring constants. Therefore, the cutoff frequency alone is not an indicator of the mass that is needed to maximize transmission (i.e., Debye temperature ratio is a poor indicator of the efficiency of transport across an interface). In Fig. 2(c), we plot the transmission for different choices of the springs on either side of the interface and $m_1 = 3$, $m_2 = 4$ and $m_{\text{int}} = m_{\text{int,max}} = 3.5$. This supports Eq. (10) and that the mass which yields the maximum transmission is independent of the choice of k_1 and k_2 and thus cutoff frequency cannot be used alone to determine the optimum choice of m_{int}. This result assumes that the spring constants connecting the impurity mass to the materials on either side of the interface are not perturbed by the inclusion of an impurity mass.

Notice that when we maximize transmission, set $m_{\text{int}} = m_{\text{int,max}}$, we recover a transmission similar to that predicted by the AMM,

$$T_{PMM}(\omega) = \frac{4Z_1(\omega)Z_2(\omega)}{(Z_1(\omega) + Z_2(\omega))^2}, \quad (11)$$

except this transmission is frequency dependent and not limited to long wavelengths (solid red line in Fig. 2(a)). Since Eq. (11) is the AMM with a substitution of phonon impedances for acoustic impedances, we have termed this transmission the PMM. Note the peak in transmission in Fig. 2(a). This peak occurs when $Z_1 = Z_2$, so that the frequency where the PMM predicts a transmission of one is

$$\omega_{Z_1=Z_2} = 2\sqrt{\frac{k_1 m_1 - k_2 m_2}{m_1^2 - m_2^2}}. \quad (12)$$

Note that there is no peak in the transmission above $\omega = 0$ when Eq. (12) is imaginary. So a peak in the transmission

will only be observed when both $Z_2(0) > Z_1(0)$ and $m_2 > m_1$ or when both $Z_1(0) > Z_2(0)$ and $m_1 > m_2$ and when $\omega_{Z_1=Z_2}$ is less than the cutoff frequency of the softer material.

Typically, thermal interface models such as the AMM and the DMM neglect the properties of the interface when predicting interface transmission, so that the predicted transmission is solely dependent on the bulk materials comprising the interface. However, a bulk semi-infinite control volume can only be constructed in two ways, either terminated by masses or by springs. Therefore, the two control volumes must be connected via either a spring or mass at the interface.

We have just discussed the case of the impurity mass and have shown that this connecting mass will result in a reduced transmission across the interface, i.e., reduce thermal boundary conduction, unless the connecting mass equals the arithmetic mean of the masses on either side of the interface. In the case that $m_{\text{int}} = (m_1 + m_2)/2$, we recover the transmission similar to the AMM transmission, which we call the PMM due to the frequency dependence and implicit sinusoidal dispersion. Thus, we satisfy the main assumption of most interface models; that the control volumes are brought together in such a way as to create a perfect or an abrupt interface. However, if m_{int} in Eq. (8) is set to either m_1 or m_2, the transmission is still reduced below the PMM prediction. Therefore, we suggest that an abrupt interface can only be obtained by making an interface where we have selected the connecting mass such that the transmission for all frequencies is solely dependent on the properties of the bulk materials.

IV. TWO ATOM CHANNEL TRANSMISSION WITH INTERFACE SPRING

We have previously discussed that two control volumes can only be coupled using a single element in two ways. In Sec. IV, we investigated how coupling with a mass affects phonon transmission. In this section, we derive the transmission for a one-dimensional system with a variable spring between the two masses as shown in Fig. 3. Following the procedure outlined in Sec. II, we find that the phonon transmission for an interface coupled by a spring is

$$T(\omega, k_{\text{int}}) = \frac{4\Gamma_1\Gamma_2}{(\Gamma_1 + \Gamma_2)^2 + 4m_1 m_2 k_1 k_2 \omega^4 \left(\frac{1}{k_{\text{int}}} - \frac{k_1+k_2}{2k_1 k_2}\right)^2 + \omega^4 \frac{(m_1 k_2 - m_2 k_1)(m_1 k_1 - m_2 k_2)}{k_1 k_2}}. \quad (13)$$

We can also rewrite this equation in terms of a modified frequency broadening function to more easily show the dependence on k_{int} as

$$T(\omega, k_{\text{int}}) = \frac{4\Gamma_1^{hs}\Gamma_2^{hs}}{(\Gamma_1^{hs} + \Gamma_2^{hs})^2 + \frac{1}{4}(\Gamma_1^{hs}\Gamma_2^{hs})^2 \left(\frac{1}{k_{\text{int}}} - \frac{k_1+k_2}{2k_1 k_2}\right)^2}, \quad (14)$$

where $\Gamma^{hs} = 4\omega^2 km/\Gamma$.

Zhang et al.[15] investigated the case of a single interface spring in a two atom channel using boundary scattering and NEGF methods and found that the maximum transmission occurs when the interface spring is the harmonic mean of the spring constants on either side of the interface,

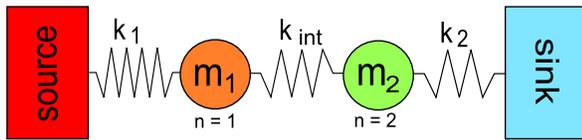$$k_{\text{int,max}} = \frac{2k_1 k_2}{k_1 + k_2}, \quad (15)$$

FIG. 3. Model system used to calculate the transmission across the interface between two dissimilar crystalline materials with variable interface adhesion. Mass, spring constant, and frequency are normalized by 1.6710^{-27} kg, 1.6710^{-3} N/m, and THz, respectively.

which is consistent with the maximum transmission as written in Eq. (13). We reproduce some of Zhang's results here and expand on them.

We plot Eq. (13) versus frequency for several values of k_{int} in Fig. 4(a) and transmission versus k_{int} for several frequencies in Fig. 4(b). For both plots $k_1 = 4$, $k_2 = 1$, $m_1 = 2$, and $m_2 = 2$ so that $k_{int,max} = 1.6$. We see in Fig. 2(a) that as k_{int} approaches $k_{int,max}$ all frequencies are maximized. We note that transmission is not symmetric about $k_{int,max}$,
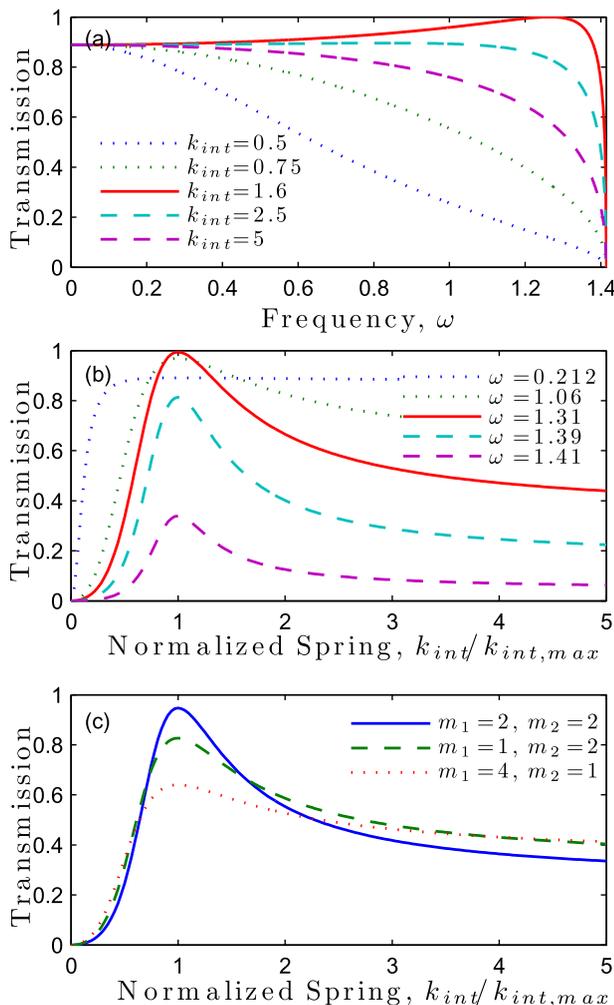

FIG. 4. Predicted transmission calculated using Eq. (13) using $k_1 = 4$, $k_2 = 1$, $m_1 = 2$, $m_2 = 2$ and $k_{int,max} = 1.6$. Mass, spring constant, and frequency are normalized by 1.6710^{-27} kg, 1.6710^{-3} N/m, and THz, respectively. (a) Plotted versus frequency for various values of k_{int}. The dashed lines indicate values of k_{int} greater than 1.6 and dotted lines for values lower than 1.6. (b) Plotted versus k_{int} for various frequencies. A maximum transmission occurs for all frequencies when $k_{int} = 1.6$. (c) Transmission versus normalized mass for $\omega = 1.36$ and various choices of m_1 and m_2. The interface spring constant which yields maximum transmission is independent of the masses.

contrasting the single atom channel case, where transmission is symmetric about $m_{int,max}$. However, if we were to plot Eq. (13) versus $1/k_{int}$, we would see the same symmetry and trends. We also notice that low frequency transmissions are not affected by bonding at the interface. This follows the same argument as for the interface mass case and if we take the limit of Eq. (13) as $\omega \rightarrow 0$, the result is Eq. (9), the low frequency transmission predicted by the AMM. The interface spring will affect low frequency transmissions, however, as k_{int} approaches zero since $k_{int} = 0$ decouples the two control volumes.

Note that the result of a spring constant other than infinity which maximizes transmission seems to be inconsistent with the results found by Shen et al.[11] Shen applied pressure to a molecular dynamics simulation cell composed of dissimilar materials to increase the adhesion at the interface. They showed that as pressure is increased, effectively increasing adhesion, thermal boundary conductance increased, which plateaued as the pressure increased past some value. However, pressures were not increased high enough to cause the bonding at the interface to increase higher than the bonding within the bulk materials. Therefore, a peak in conductance might not have fallen within the investigated parameter space. They used NEGF to explain their results by showing that the phonon transmission across a 1D analogue of there MD simulation also plateaued after the bond strength of the interface reached a certain value. Their phonon transmission results are actually consistent with our calculations, but their calculations were done for low frequency, $\sim\omega_{c,1}/4$, so that the peak transmission was not observed. This plateau effect can also be seen in our results, Fig. 4(b), at low frequency. On the other hand, the inconsistencies might be due to the limits of our model; harmonic, nearest neighbor interactions, and one-dimensional. Anharmonic scattering combined with three-dimensional densities of states may add additional scattering mechanisms at the interface which could lead to the observed plateau of the conductance.

In Fig. 4(b), we see more clearly that high frequency phonons are affected more than low frequency phonons by the interface bonding. We note that as k_{int} goes below $k_{int,max}$ the transmission falls quickly to zero. However, as k_{int} increases above $k_{int,max}$, the transmission decreases asymptotically to a frequency dependent value. It can be shown that this transmission is identical to the case where m_{int} in Eq. (8) is set to zero. This implies that there are four cases that result in identical transmissions; $k_{int} = k_1 k_2/(k_1 + k_2)$, $k_{int} = \infty$, $m_{int} = m_1 + m_2$ and $m_{int} = 0$. The case where $k_{int} = k_1 k_2/(k_1 + k_2)$ is identical to the case where $m_{int} = 0$ because having a zero mass is like adding the two springs on either side of the mass in series, resulting in an effective spring with spring constant $k_1 k_2/(k_1 + k_2)$. In an analogous manner, the case where $k_{int} = \infty$ is identical to $m_{int} = m_1 + m_2$ because an infinitely strong spring effectively fuses the two masses flanking it together resulting in an effective mass of $m_1 + m_2$. This then results in an impurity mass which reduces the transmission of phonons across the interface. Each of the four cases can be shown to be equivalent mathematically using Eqs. (8) and (13).

Since there is only one term in the denominator which is dependent on k_{int}, and since this term is always positive,

minimizing it will always maximize transmission for all frequencies. Similar to the first case, the interface spring which maximizes the transmission is only dependent on the spring constants on either side of the interface and is not dependent on frequency or mass as is shown in Figs. 4(b) and 4(c). So, this choice of interface spring constant maximizes phonon transmission for all frequencies. However, we also notice that there are three terms in the denominator as opposed to two in the interface mass case, Eq. (8). The first two in Eq. (13) are similar to the two in the transmission for the interface mass case, where the first is due to the mismatch in phonon impedances, and second term is due to the interface impurity. However, for the interface spring case, Eq. (13) contains an additional term which is not dependent on the impurity, but only on the spring constants and masses of the two sides of the interface. Therefore, even when k_{int} equals the harmonic mean of the spring constants, we do not recover the PMM transmission, Eq. (11), rather the PMM transmission with some perturbation term which is independent of the coupling spring. We will define this perturbation term without the factor of ω^4 as Δ, so that

$$\Delta = \frac{(m_1 k_2 - m_2 k_1)(m_1 k_1 - m_2 k_2)}{4 k_1 k_2} \qquad (16)$$

$$= (Z_2^2(0) - Z_1^2(0)) \left(\frac{\omega_{c,1}^2 - \omega_{c,2}^2}{\omega_{c,1}^2 \omega_{c,2}^2} \right). \qquad (17)$$

This term goes to zero when either the acoustic impedances, $Z(0) = \sqrt{km}$, are equal or when the cutoff frequencies, $\omega_c = 2\sqrt{k/m}$, are equal. This means that if $\Delta = 0$, the phonon impedances only equal at one of the extremes of the frequency range, i.e., when $\omega = 0$ or $\omega = \omega_c$. Also, note that this term can be both positive or negative whereas the rest of the terms can only be positive. This implies that Δ can either decrease the transmission for all frequencies, $\Delta > 0$, or increase the transmission for all frequencies, $\Delta < 0$ relative to the PMM. Recall that in the interface mass case, the PMM is an upper bound for transmission. However, an interface spring has the ability to increase the transmission above that of the PMM. Therefore the Δ term can be thought of as a term resulting from resonance at the interface due to the finite length of the coupling spring, which results in either an increased or decreased transmission when compared to the Eq. (11). A resonance, constructive interference, only occurs when there does not exist a frequency in which $Z_1(\omega) = Z_2(\omega)$ and an anti-resonance, destructive interference, occurs only when $Z_1(\omega) = Z_2(\omega)$. Note that when $k_1 = k_2$, $\Delta \geq 0$, resulting in a reduced transmission, i.e., anti-resonance. However, when $m_1 = m_2$, $\Delta \leq 0$, resulting in an increased transmission, i.e., resonance. It should be mentioned that Δ does not cause additional resonance, rather it shifts the frequency of maximum transmission or maximum resonance resulting in an increased or decreased transmission when compared to the PMM as will be shown in Sec. V.

As we discussed in Sec. III, when two control volumes are coupled via an interface mass equal to the arithmetic mean of the masses on either side of the interface a perfect or abrupt interface results. Under this condition, we satisfy the main assumption of the majority of thermal interface models; that the transmission of phonons is dependent solely on the bulk properties of the materials comprising the interface. In this section, we investigated the case of coupling two control volumes with an interface spring. We found that when we set k_{int} equal to the harmonic mean of the springs on either side of the interface, we maximize transmission. Since Eq. (13) is only dependent on the properties of the source and sink when $k_{int} = k_{int,max}$, one might think that we have created an abrupt interface. In some sense we have, since we have removed any scattering at the interface due to the impurity spring. However, due to the finite length of the coupling spring an additional term in the transmission exists. Therefore, an abrupt interface cannot be created for the interface spring case in exactly the same sense as the impurity mass case.

V. COMPARING MAXIMUM TRANSMISSIONS

In Sec. IV, we showed that coupling with a spring results in a term in the transmission equation due to resonance at the interface. In this section, we investigate the effect of this resonance term with respect to the interface mass case, when $k_{int} = k_{int,max}$ and $m_{int} = m_{int,max}$. We also investigate the conditions when coupling with a spring is equivalent to coupling with a mass.

We can see how coupling with a spring augments the transmission when compared to the PMM, Eq. (11), by rewriting Eq. (13) with $k_{int} = k_{int,max}$ as

$$T(\omega) = \frac{4 \frac{\Gamma_1}{k_1 m_1} \frac{\Gamma_2}{k_2 m_2}}{\left(\frac{\Gamma_1}{k_1 m_1} + \frac{\Gamma_2}{k_2 m_2} \right)^2} = \frac{4 \Gamma_1^{hs} \Gamma_2^{hs}}{(\Gamma_1^{hs} + \Gamma_2^{hs})^2}. \qquad (18)$$

In this form, it is easy to see that in order to maximize transmission with $k_{int} = k_{int,max}$, we must set $\frac{\Gamma_1}{m_1 k_1} = \frac{\Gamma_2}{m_2 k_2}$. Therefore, when coupling two control volumes with an interface spring, we do not match phonon impedances to maximize transmission as predicted by the PMM, instead we match $Z(\omega)/Z^2(0)$. This means that the location of the maximum transmission will be shifted from what is predicted by Eq. (12) for the PMM to,

$$\omega_{max} = 2 \sqrt{\frac{k_1 k_2 (m_2 k_2 - m_1 k_1)}{m_1 m_2 (k_2^2 - k_1^2)}}. \qquad (19)$$

Therefore, when the PMM or Eq. (12) predicts no peak in the transmission, Δ may shift the transmission such that a peak is observed or vice versa. The shift in the resonant frequency caused by Δ results in an increased or decreased transmission when compared to the PMM.

To demonstrate this point, we have plotted the transmission predicted by the PMM, Eq. (11), and the modified PMM, Eq. (18), versus frequency in Fig. 5(a). We have chosen to set $k_{int} = k_{int,max}$ in Eq. (13) to simplify the results and show only the effect of the resonant term, Δ, on the prediction of the PMM. The addition of the k_{int} dependent term will only decrease the transmission over all frequencies since
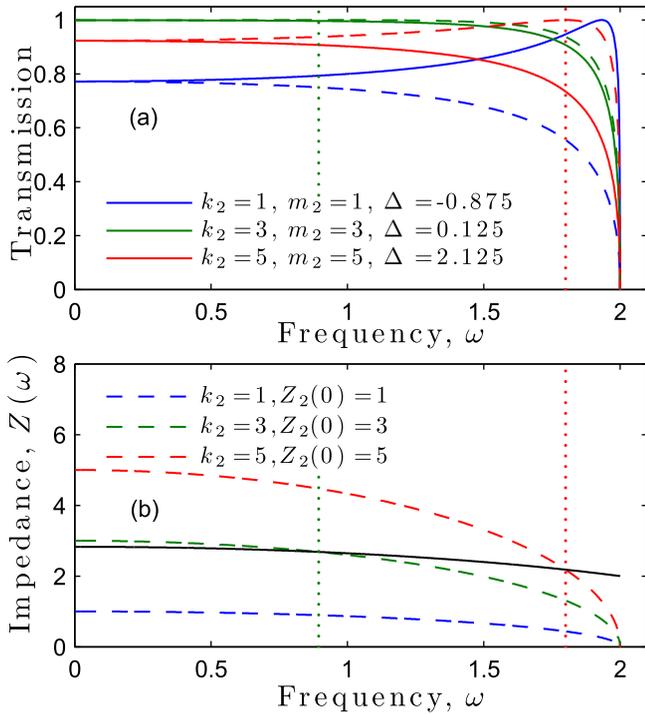
FIG. 5. (a) Transmission predicted by the PMM (dashed line-Eq. (11)), and by our modified PMM (solid line-Eq. (18)). Mass, spring constant, and frequency are normalized by $1.67 10^{-27}$ kg, $1.67 10^{-3}$ N/m, and THz, respectively. Depending on the differences in the low frequency acoustic impedances, the NEGF transmission can be increased or decreased when compared to the PMM. The vertical lines indicate the frequency where the acoustic impedances are equal. (b) Frequency dependent acoustic impedances, Eq. (6), of side 1 (solid line) and side 2 (dashed lines) versus frequency. When the impedance of side 1 matches side 2, the PMM transmission is maximized to a value of 1. However, this is not the case when the control volumes are coupled with a spring (solid lines), Eq. (18).

it is always positive. The properties of side one remain constant, $m_1 = 2$ and $k_1 = 4$, and we vary m_2 and k_2 while keeping $\omega_{c,2} = 2$. The solid lines are the transmission predicted by Eq. (18), while the dashed line of the same color is the PMM prediction and the vertical dotted line of the same color is the frequency where $Z_1 = Z_2$. The value of Δ is shown in the legend. We also plot the acoustic impedances versus frequency in Fig. 5(b). The solid black line is the acoustic impedance of side 1, the dashed lines are acoustic impedances of side 2 where the vertical line of the same color is the frequency at which $Z_1 = Z_2$.

We begin with the first case, $m_2 = k_2 = 1$. First, notice that there is no peak in the PMM transmission at high frequencies. This is because the phonon impedances are never equal as can be seen in Fig. 5(b). However, if we include Δ, in this case $\Delta = -0.88$, the transmission is increased over all frequencies resulting in a peak in the transmission near the cutoff frequency. The reason for this peak, is that $\Gamma_1/k_1 m_1 = \Gamma_2/k_2 m_2$ at the frequency of the peak. Note that Γ_1 does not intersect Γ_2 if $\Gamma_1/k_1 m_1$ intersects $\Gamma_2/k_2 m_2$ and vice versa, unless $\Delta = 0$. In the next case, $k_2 = 5$ and $m_2 = 5$, we see that the PMM predicts a peak in the transmission near the cutoff frequency since the phonon impedances are equal near $\omega = 1.8$. However, due to Δ being greater than zero, the transmission is decreased for all frequencies so that we no longer see the peak in transmission. For the third case,

$k_2 = 3$ and $m_2 = 3$, there is little difference between the transmissions. This is due to the acoustic impedances being nearly matched, $Z_1(0) = \sqrt{8}$ and $Z_2(0) = \sqrt{9}$, leading to a small value of Δ and good agreement between the transmissions. Finally notice that at low frequencies all cases become equivalent to the PMM. As has been discussed, in the limit of low frequencies, the transmission in this case, Eq. (13), becomes the low frequency AMM transmission, Eq. (9).

Even though these two model systems predict different transmissions at high frequencies, if $\Delta < 0$, we may choose k_{int} such that we recover the AMM when,

$$\frac{1}{k_{int,PMM}} = \frac{k_1 + k_2}{2k_1 k_2} \pm \sqrt{\left(\frac{k_1 + k_2}{2k_1 k_2}\right)^2 - \frac{(m_1 + m_1)^2}{4k_1 k_2 m_1 m_2}} \quad (20)$$

$$= \frac{1}{k_{int,max}} \pm \sqrt{\frac{-\Delta}{Z_1^2(0)Z_2^2(0)}}. \quad (21)$$

Notice that there are two choices of k_{int} which recover the PMM, one less and one greater than $k_{int, max}$. It can be shown that when $m_1 = m_2$, the interface spring constant which recovers the PMM is either k_1 or k_2. This is because under this condition, we recover the interface mass case with the interface mass set to the arithmetic mean.

Since coupling with a spring and a mass yield different results for the phonon transmission even when the transmissions are maximized, we need to know when they are equivalent. If we set the last two terms in the denominator of the interface spring transmission, Eq. (13), equal to the second term in the denominator of the interface mass transmission, Eq. (8), we are able to find the equivalent m_{int} for a choice of k_{int} as

$$m_{int} = m_{int,max} \pm \sqrt{m_1 k_1 m_2 k_2 \left(\frac{1}{k_{int}} - \frac{1}{k_{int,max}}\right)^2 + \Delta}. \quad (22)$$

When k_{int} is close to $k_{int,max}$ and Δ is negative the equivalent m_{int} may be imaginary, meaning that no choice of the interface mass will be equivalent to the coupling using an interface spring. This is because under certain circumstances coupling with a spring can result in a transmission greater than that of the PMM, whereas the maximum transmission possible when coupling with a mass is the PMM. Therefore in some cases, there is no m_{int} which is equivalent to a k_{int}.

VI. CONCLUSION

Recently, interface properties have garnered much interest for tuning thermal boundary conductance in nanostructured devices. Many studies have demonstrated the ability to tune thermal conductivity via carefully controlling the properties of the interface both in theory[7,11,15,17,18] and experiment.[1–5,19] However, many of the studies have not been able to separate the effect each property of the interface. In order to illustrate the individual effects of bonding and impurity mass on thermal transport, we derived analytical equations for the transmission of phonons in an one-dimensional

harmonic system and investigated how each property effects the predicted transmission.

The two case considered were a single mass sitting at the interface between two semiinfinite control volumes and two control volumes coupled by a variable interatomic bond. In the interface mass case, we showed that it is possible to maximize transmission for all frequencies by setting the interface mass equal to the arithmetic mean of the masses on either side of the interface. When we do this, not only do we maximize transmission but we recover the transmission predicted by the frequency dependent AMM, the PMM. Therefore, the maximum transmission that can be obtained by coupling two control volumes with a mass at the interface is the PMM transmission. In the case of the interfacial spring, we showed that we can maximize the transmission by setting the interface spring to the harmonic mean of the springs on either side of the interface. However, in this case the maximum transmission is not necessarily the transmission predicted by the PMM, rather it is the PMM with an additional resonance term which can increase or decrease the transmission relative to the PMM transmission. This means that in some cases, the maximum transmission depends on whether the control volumes are connected by a mass or a spring and that an impurity spring at the interface in some situations can increase transmission above that when no impurity is observed at the interface. This implies that setting the interface spring equal to the harmonic mean does not necessarily create an abrupt interface as defined when the interface mass is set to the arithmetic mean. Therefore, coupling two control volumes with a spring and a mass and maximizing the transmission in each case will not necessarily result in the same thermal boundary conductance.

ACKNOWLEDGMENTS

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