Effect of electron-phonon coupling on thermal transport across metal-nonmetal interface — A second look

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Abstract – The effect of electron-phonon (e-ph) coupling on thermal transport across metal-nonmetal interfaces is yet to be completely understood. In this paper, we use a series of molecular-dynamics (MD) simulations with e-ph coupling effect included by Langevin dynamics to calculate the thermal conductance at a model metal-nonmetal interface. It is found that while e-ph coupling can present additional thermal resistance on top of the phonon-phonon thermal resistance, it can also make the phonon-phonon thermal conductance larger than the pure phonon transport case. This is because the e-ph interaction can disturb the phonon subsystem and enhance the energy communication between different phonon modes inside the metal. This facilitates redistributing phonon energy into modes that can more easily transfer energy across the interfaces. Compared to the pure phonon thermal conduction, the total thermal conductance with e-ph coupling effect can become either smaller or larger depending on the coupling factor. This result helps clarify the role of e-ph coupling in thermal transport across metal-nonmetal interface.

Introduction. – Thermal transport across metal-nonmetal interfaces is of great importance for applications such as microelectronics, thermoelectrics and electro-optical devices [1,2]. In metals, electrons and phonons can both transfer heat, while in nonmetals thermal transport is exclusively dominated by phonons. Since phonons are the common heat carriers in metals and nonmetals, it is generally accepted that thermal transport across their interfaces is mediated by phonon-phonon coupling [3–5]. However, electron and phonon in metals are not isolated but interacting through electron-phonon (e-ph) coupling, enabling energy exchange between them. Whether such coupling is important for interfacial thermal transport is still under debate.

Majumdar and Reddy [3] used a two-temperature model to explain the role of e-ph coupling in metal as a thermal resistance between electron and phonon subsystems. This adds additional resistance to the intrinsic phonon-phonon interfacial resistance. However, Singh et al. [4] recently used the Bloch-Boltzmann-Peierls formula [6] and concluded that the e-ph coupling does not contribute significantly to the interfacial thermal conductance. One effect that has not been considered in these models is that the inelastic scattering between electron and phonon can perturb the phonon subsystem and can potentially result in a re-distribution of the phonon energy in the metal. For example, the usual e-ph scattering processes at room temperature involve one electron converting from one state to another with the absorption or emission of a phonon [7]. Higher-order inelastic scatterings involving multiple electrons and phonons are also possible depending on the specific materials and temperature. These effects will change the populations of the phonons involved in the e-ph scattering processes and thus influence the scattering within the phonon subsystem [8–10]. Such an influence in the energy exchange among phonons can potentially lead to better phonon-phonon interfacial thermal transport [11]. As a result, e-ph coupling can have two competing effects: it can add additional resistance on top of the resistance from interfacial phonon reflection, but it can also improve the phonon-phonon conductance across the interface due to its perturbation effect on the phonon subsystem.

The two-temperature model has been incorporated into molecular-dynamics (MD) simulations using Langevin

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dynamics to simulate the effect of e-ph coupling on the phonon system (e.g., atomic trajectory) [12]. Such a method should be able to capture the aforementioned effect of e-ph coupling on the phonon transport across interfaces. However, different conclusions were obtained in the literature. Wang et al. [13] used the two-temperature MD to calculate the thermal conductance at a silicon-copper interface. It is observed that by including the e-ph effect, interfacial thermal conductance is significantly reduced. Jones et al. [14] and Nuo et al. [15] simulated the metal-nonmetal interface using a similar method. However, the e-ph coupling effect is found negligible for the interfacial thermal transport.

In this paper, we revisit the e-ph coupling effect on thermal transport across metal-nonmetal interfaces. From the two-temperature model, we first derive a general expression of total thermal conductance as a function of e-ph coupling factor, thermal conductivities of electrons and phonons, phonon-phonon conductance. We find that the Majumdar-Reddy serial resistance model is a special case of the derived general formula. By performing MD simulation with the two-temperature model and interpreting the data using the derived model, we find that e-ph coupling can perturb the phonon subsystem and increase the data using the derived model, we find that e-ph coupling effect is found negligible for the interfacial thermal transport.

Theory. – Majumdar and Reddy [3] first introduced the two-temperature model to explain the role of e-ph coupling in thermal conductance of a metal-nonmetal interface. Their derived model reads

\[ h_{\text{total}} = \frac{h_{pp} \cdot \sqrt{g \cdot k_p}}{h_{pp} + \sqrt{g \cdot k_p}}. \]

where \( h_{pp} \) is the phonon-phonon thermal conductance at the interface, \( g \) is the e-ph coupling factor and \( k_p \) is the phonon thermal conductivity of the metal. Such an expression was obtained by solving the following coupled equations for electron and phonon subsystems in the metal side, respectively:

\[ k_e \frac{d^2 T_e(x)}{dx^2} - g \cdot (T_e(x) - T_p(x)) = 0, \]  
\[ k_p \frac{d^2 T_p(x)}{dx^2} + g \cdot (T_e(x) - T_p(x)) = 0. \]

The solutions to these equations are in the following forms (see fig. 1):

\[ T_e(x) = A \cdot x + B + C \cdot \exp \left( \frac{x}{T} \right) + D \cdot \exp \left( -\frac{x}{T} \right), \]  
\[ T_p(x) = A \cdot x + B - C \cdot \frac{k_e}{k_p} \cdot \exp \left( \frac{x}{T} \right) - D \cdot \frac{k_e}{k_p} \cdot \exp \left( -\frac{x}{T} \right), \]

where \( k_e \) is electronic thermal conductivity, and \( A, B, C \) and \( D \) are constants. \( l = \sqrt{\frac{k_e k_p}{g(k_p + k_e)}} \) can be regarded as a characteristic length of e-ph coupling.

Below we show the derivation of the formulation of the interfacial thermal conductance at the metal-nonmetal interface. Fourier’s law results in a linear profile of the total temperature of metal (right-hand side) and the temperature of nonmetal (left-hand side) at either side of the interface (fig. 1):

\[ T_{\text{right}}(x) = A \cdot x + B, \]  
\[ T_{\text{left}}(x) = E \cdot x + F, \]

where \( A, B, E \) and \( F \) are constants. The boundary conditions and the conditions at the interface are listed below and are indicated in fig. 1. It is worth noting that we have assumed that the electrons do not transfer heat across the interface, which is consistent with the argument in the Majumdar-Reddy model [3]:

\[ T_p(L) = T_e(L) = T_{\text{high}}, \]  
\[ T_{\text{left}}(-L_{\text{left}}) = T_{\text{low}}, \]  
\[ \frac{dT_e(x)}{dx} \bigg|_{x=0} = 0, \]  
\[ -k_p \frac{dT_p(x)}{dx} \bigg|_{x=0} = -k_{\text{left}} \frac{dT_{\text{left}}(x)}{dx} = h_{pp} \cdot (T_p(0) - T_{\text{left}}(0)), \]

where \( L \) is the thickness of the right-hand side material (metal) and \( L_{\text{left}} \) is that of the left-hand side material (nonmetal). Equation (5d) also indicates how the phonon-phonon interfacial thermal conductance, \( h_{pp} \), is defined. Then the total thermal conductance at interface is defined as the ratio of total heat flux to the temperature difference at the interface (note here that \( T_{\text{right}} \) refers to the total temperature of the metal):

\[ h_{\text{total}} = \frac{-k_{\text{left}} \frac{dT_{\text{left}}(x)}{dx}}{T_{\text{right}}(0) - T_{\text{left}}(0)}. \]

Combining eqs. (4), (5) and (6), we can express the total thermal conductance, \( h_{\text{total}} \), as a function of \( k_e, k_p, g, h_{pp} \)
and $L$, we can derive the total interfacial thermal conductance:

$$h_{\text{total}} = \frac{k_p \cdot h_{pp} \cdot (k_e + k_p) \cdot \cosh \left( \frac{L}{l} \right)}{h_{pp} \cdot k_e \cdot l \cdot \sinh \left( \frac{L}{l} \right) + k_p \cdot (k_e + k_p) \cdot \cosh \left( \frac{L}{l} \right)}.$$  \hspace{1cm} (7)

It is also worth noting that when $k_e = 0$ (e.g., electron is not contributing to heat transfer), $h_{\text{total}} = h_{pp}$. If we further apply the condition that $k_e \gg k_p$ and at thick metal limit $L/l \gg 1$, the total thermal conductance will further deduce to the Majumdar-Reddy model (eq. (1)) [3]. It can be seen from eq. (7) that the total thermal conductance will always be smaller than phonon-phonon thermal conductance. However, the phonon-phonon thermal conductance, $h_{pp}$, itself should be a function of coupling factor, $g$, according to the aforementioned reason: e-ph interaction can perturb the phonon subsystem and redistribute energy among different phonons and thus facilitate interfacial phonon-phonon energy transport. Phenomenologically, it is also easy to understand that the actual phonon temperature profile in metal and at the interface will be different under the same heat flux condition whether the e-ph interaction is present or not. As a result, due to this effect, it is possible that the total thermal conductance can be larger than the intrinsic phonon-phonon conductance. When the Majumdar-Reddy’s model is used, such an effect was usually ignored, and $h_{pp}$ was usually obtained from mismatch models [16,17]. These models are developed for phonon-phonon interfacial thermal conductance without the consideration of e-ph coupling effects.

In the rest of this paper, we show through MD simulations that $h_{pp}$ will increase as we increase the coupling factor $g$. Compared to the pure phonon heat transfer case, we find that the total thermal conductance including e-ph coupling effect can indeed become either smaller or larger, depending on the e-ph coupling factor $g$.

**Results and discussions.** – MD simulations [18] are performed on a model bi-material junction as shown in fig. 2(a), and Langevin dynamics [19] is included for the atomic motion with the friction force and the stochastic force representing the effects of energy communication between electron and phonon subsystems. The detail of the simulation can be found in the supplemental information [20].

We first study a case without e-ph interaction to obtain the intrinsic phonon-phonon thermal conductance, $h_{pp,\text{intrinsic}}$. The total thermal conductance is calculated as $h_{pp,\text{intrinsic}} = J/\Delta T$, where $J$ is heat flux crossing the interface which can be obtained by calculating the energies added to or taken out from the heat baths, and $\Delta T$ is obtained by extrapolating the linear portion of the temperature profiles in the materials and take the difference at the interface (fig. 2(b)). We then performed MD simulations with e-ph interactions. Different e-ph coupling factors, $g$, and electronic thermal conductivity, $k_e$, are used. In these simulations, we strive to obtain two thermal conductances: $h_{\text{total}}$ and $h_{pp}$. Extracting $h_{\text{total}}$ is similar to how we extract $h_{pp,\text{intrinsic}}$. The only difference is that for the metal side, the linear extrapolation was done on the total temperature profile. However, the same method is not applicable to extracting $h_{pp}$ since the phonon temperature is not linear due to the e-ph coupling (eq. (3b)). To obtain $h_{pp}$, we utilize eq. (3) to fit the electron and phonon temperature profiles away from the interface. Fitting temperature profiles away from the interface is necessary since the nonlinearity of the phonon temperature near the interface is not due to e-ph coupling but rather the fact that atoms near the interface has difference vibrational feature from those inside the material and thus present additional resistance. This is because atoms near the interface will experience different force constants from those inside the material. Our theoretical model does not consider these effects. However, away from the interface, our model is valid. The fitted curve is extended to the interface and $h_{pp}$ is obtained utilizing the temperature difference of extrapolated phonon temperature profiles at the interface (see fig. 2(c)). Using such a treatment, we have effective...
lumped the above-mentioned temperature bending effect into the interfacial thermal conductance data.

The interfacial thermal conductance as a function of electron-phonon coupling factor, $g$, and the electron thermal conductivity, $k_e$, are presented in fig. 3. Electron thermal conductivity values chosen are 0.1, 1 and 10 W/mK. These values are respectively much smaller than, on the same order of magnitude as, and much larger than the phonon thermal conductivity of the lattice ($\sim$ 1 W/mK) [11], roughly covering the range of highly doped semiconductors [7], metallic carbon nanotube [21] to common noble metals [13]. For the e-ph coupling factor $g$, we choose values from $0.1 \times 10^{17}$ to $5 \times 10^{17}$ W/m$^3$K covers many common materials [22] (e.g., Ag: $0.2 \times 10^{17}$; Al: $2.45 \times 10^{17}$; Pt: $5 \times 10^{17}$ W/m$^3$K). It should be pointed out that the referred values are room temperature coupling factors. However, since MD simulations are classical, the simulations in this study, even though performed at a mean temperature of 15 K, actually correspond to cases well above the Debye temperature. For most metals, the room temperature approaches this limit since they usually have relatively low Debye temperatures (e.g., Ag: 215 K; Al: 428 K; Pt: 240 K). As a result, the use of room temperature coupling factor should still be relevant. It is also worth noting that there are some $h_{pp}$ points missing for the $k_e = 0.1$ W/mK cases. It is because for these cases (e.g., small $k_e$ and large $g$), the fittings were not successful. Nevertheless, the available data are sufficient to support our following argument.

Figure 3(a) presents data for cases with the e-ph interaction applied to the monatomic lattice on the left-hand side of the interface. These are common cases since most metals are monatomic. As seen from fig. 3(a), the total thermal conductance, $h_{total}$, increases monotonically when $g$ increases. Compared to the intrinsic phonon-phonon conductance ($h_{pp,intrinsic}$, dotted horizontal line), $h_{total}$ can be either smaller or larger, depending on the electron thermal conductivity and coupling factor. We attribute such an observation to the competing effects e-ph coupling has on the interfacial thermal conductance, i.e., it adds an additional thermal resistance serially to $h_{pp}$ but can also enhance $h_{pp}$ itself due to its perturbation effect on the phonon system. This is proven in fig. 3 which shows that $h_{pp}$ increases monotonically with $g$. It is also seen that $h_{total}$ is smaller than $h_{pp}$ in all cases. This agrees with eq. (7) which indicates that e-ph coupling always add additional resistance to $h_{pp}$. Another observation worth mentioning is that the difference between $h_{pp}$ and $h_{total}$ becomes smaller as $g$ becomes larger. This is because larger $g$ facilitates the energy communication between electrons and phonons, and thus the resistance due to their coupling effects becomes smaller.

We have also simulated cases with the e-ph interaction applied to the diatomic lattice on the right-hand side of the interface. Such simulations are also of practical relevance since materials like NbSe$_2$ and metallic carbon nanotube [21] are multi-atomic metals, and highly doped semiconductors can also have non-negligible electron contribution in heat transfer [7]. As shown in fig. 3(b), the trend of $h_{total}$ and $h_{pp}$ from these cases are the same as those in the previous cases shown in fig. 3(a). These cases also help prove that the perturbation due to e-ph coupling can enhance the energy communication between different phonon modes in metal. Such an effect was visualized by calculating the spectral temperatures of the optical and acoustic phonons in the diatomic lattice. The spectral temperature can be utilized to characterize how equilibrated different phonons are in terms of energy communication [23–25]: the closer the spectral temperatures, the better the energy equilibration. The details of the calculation can be found in ref. [9]. It is found from fig. 4 that larger e-ph coupling factor will increase the energy interaction between optical and acoustic phonons as indicated by the closer spectral temperatures of these two groups of

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modes. We have previously shown that such an enhanced internal energy communication will increase phonon thermal conductance at interface [11].

Comparing fig. 3(a) and (b), we found that for the cases in fig. 3(b) $h_{\text{total}}$'s are rarely smaller than $h_{\text{pp, intrinsic}}$. The only difference between these two sets of cases is that in fig. 3(b) the e-ph coupling effect is applied to the diatomic lattice. According to our previous study [11], the interactions between optical and acoustic phonons can greatly influence the acoustic phonon transport across the interface. When e-ph coupling effect is applied to the diatomic lattice, such interactions can be enhanced. When e-ph coupling effect is applied to the monatomic lattice, which only has acoustic phonons, the interactions within acoustic modes are enhanced. While energy exchange within acoustic phonons can also help redistributing energy to improve thermal transport across the interface, it is possible that the energy exchange between acoustic and optical phonons has a stronger effect on the interfacial thermal conductance. This could be the reason why we see a more pronounced enhancement effect of e-ph coupling on $h_{\text{total}}$ in fig. 3(b).

It is also notable that for a given e-ph coupling factor, $g$, the interfacial thermal conductance will increase as electron thermal conductivity, $k_e$, decreases. When $k_e$ is low (0.1 W/mK, red curves, fig. 3), the dominant heat carriers are phonons. In this case, the perturbation effect of the e-ph coupling is more dominant. Such a perturbation to the phononic system will enhance the phonon-phonon interfacial thermal conductance. When the coupling factor is large, such perturbation effect is stronger, and thus we see an increase in the phonon-phonon thermal conductance (fig. 3).

On the other hand, when electron thermal conductivity is high ($k_e = 10$ W/mK, green curves, fig. 3), the electron will dominate the heat transfer. In this case, electrons need to transfer energy to phonons which then carry energy across the interface. The resistance role of the e-ph coupling thus dominates its effects on the interfacial thermal transport process. In these cases, the electron thermal conductivity is much larger than the phonon counterpart, and common metals like copper, aluminum and gold fall into this category. In this scenario, the Majumdar-Reddy picture [3] should be applicable.

For the previously mentioned two-temperature MD works [13–15], we can qualitatively explain the discrepancy among different simulations based on the conclusion obtained from the present study. For Wang’s work [13], copper was used, while aluminum was used in the other two studies [14,15]. Copper has a much smaller e-ph coupling factor ($g = 0.5 \times 10^{17}$ W/m$^3$K) and a larger electron thermal conductivity than those of aluminum ($g = 2.4 \times 10^{17}$ W/m$^3$K) [22], respectively. According to fig. 3(a), e-ph coupling in copper is expected to present much larger thermal resistance than aluminum, which was found from these simulations [13–15].

**Conclusion.** In this paper, we found that the phonon-phonon thermal conductance at metal-nonmetal interfaces increases as the e-ph coupling factor in the metal side increases. This is because energy communications between different phonon modes in metal are strengthened by e-ph interactions which facilitates energy redistribution into modes that can more easily transfer energy through the interfaces. It is found that the serial thermal resistance model proposed by Majumdar and Reddy is applicable under high electron thermal conductivity and small e-ph coupling situations. The total thermal conductance may be smaller or larger than the intrinsic phonon-phonon conductance depending on the actual e-ph coupling factor and thermal conductivity of electrons in real materials.

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