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The critical particle size for enhancing thermal conductivity in metal nanoparticle-polymer composites

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Polymers used as thermal interface materials are often filled with high-thermal conductivity particles to enhance the thermal performance. Here, we have combined molecular dynamics and the two-temperature model in 1D to investigate the impact of the metal filler size on the overall thermal conductivity. A critical particle size has been identified above which thermal conductivity enhancement can be achieved, caused by the interplay between high particle thermal conductivity and the added electron-phonon and phonon-phonon thermal boundary resistance brought by the particle fillers. Calculations on the SAM/Au/SAM (self-assembly-monolayer) system show a critical thickness \( L_c \) of around 10.8 nm. Based on the results, we define an effective thermal conductivity and propose a new thermal circuit analysis approach for the sandwiched metal layer that can intuitively explain simulation and experimental data. The results show that when the metal layer thickness decreases to be much smaller than the electron-phonon cooling length (or as the “thin limit”), the effective thermal conductivity is just the phonon portion, and electrons do not participate in thermal transport. As the thickness increases to the “thick limit,” the effective thermal conductivity recovers the metal bulk value. Several factors that could affect \( L_c \) are discussed, and it is discovered that the thermal conductivity, thermal boundary resistance, and the electron-phonon coupling factor are all important in controlling \( L_c \).

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I. INTRODUCTION

Nanocomposite materials are widely used nowadays due to their outstanding properties which cannot be achieved by single-phase materials. They can be implemented in conditions where a high thermal conductivity is desired for heat dissipation, or where a low thermal conductivity is desired for a large thermoelectric figure of merit \( ZT \).\(^1\)\(^-\)\(^4\) Besides, they play important roles in constructing controllable nanostructures, such as nanowires, nanotubes, and nanoparticles.\(^5\) Many practical applications require us to optimize conflicting properties of materials to meet the demands. This could often be achieved by combining materials of different properties or adding one to another. Thus, predicting the property of the composites and understanding the physics and mechanism become important. More than one hundred years ago, Maxwell already presented a theoretical method for calculating the effective properties of particulate composites.\(^6\) His pioneer work has served as a basis for many following studies through these years.

There have been a series of theoretical studies on the effective properties of composite materials. The effective medium approximation (EMA) has been most widely applied.\(^7\)\(^-\)\(^10\) Hasselman and Johnson developed a Maxwell-Garnett model based analysis for composites with different types of particle inclusions.\(^11\) It was discovered that the particle size could affect the effective thermal conductivity. Every \( et \) al. compared the Maxwell-Garnett model and Bruggeman model and discussed the effect of the particle size in the form of a length-unit parameter: Kapitza radius.\(^12\) By combining EMA with multiple scattering theory, Nan \( et \) al. developed a model for arbitrary composites where they considered the thermal boundary resistance (TBR) in the form of a coated layer.\(^13\) Duan \( et \) al. derived an explicit expression for the effective thermal conductivity of heterogeneous media containing ellipsoidal inclusions. More recently, they extended their previous study to include the effect of the imperfect bond between the inclusions and the matrix as well.\(^14\)\(^,\)\(^15\)

However, in these models, only phonons are considered, so they are not expected to work well for the metal-matrix composites (MMC) where electrons can make a significant difference.\(^16\) In more recent studies, new methods and modified approaches are developed to take into account these effects. Several works on formulating the two-temperature model (TTM)\(^17\) and Boltzmann transport equations (BTE) have already been presented.\(^18\)\(^,\)\(^19\) Combining their previous work with TTM, Miranda \( et \) al.\(^20\) derived an analysis for the effective thermal conductivity of particulate composites with oriented spheroidal metallic particles embedded in a dielectric matrix. In their following study, extended models which can account for composites in the non-dilute limit were also derived by means of the crowding factor\(^21\) and EMA.\(^22\)

The effect of particle sizes on the effective thermal conductivity has been investigated by several studies,\(^12\)\(^,\)\(^20\) but an explicit analytical solution considering electrons’ effect has not been given. The critical radius has a significant effect on the effective thermal property; therefore, developing a useful model for predicting \( r_c \) is practical for guiding us in choosing the materials and sizes of particles when synchronizing a composite material. Also, modern
time domain thermal reflectance (TDTR) experiments often involve metal-dielectric multilayers, and it will be beneficial to develop analytical relations that are easy to use. In this study, we combine molecular dynamics (MD) with TTM-Fourier calculations to predict the critical thickness \( L_c \) in 1D for metal particles embedded in a polymer matrix. A general solution to TTM that is applicable regardless of the size of the system is derived. The SAM/Au/SAM case study is presented as an example, and TTM-MD simulation is performed to predict the thermal properties for the inputs of the calculation. Based on the mathematical equations and thermal circuit analysis, we propose a new approach to analyze the thermal conduction in metal thin films sandwiched between dielectric layers, which is more intuitive in explaining experimental and simulation results than previous models under certain conditions. Finally, several factors including the thermal conductivity, TBR, and electron-phonon coupling that can affect \( L_c \) are discussed.

II. TTM-FOURIER APPROACH

Generally, metals have higher thermal conductivity than polymers. Therefore, when metal particles are added to the polymer matrix, the thermal resistance is expected to be reduced. However, TBR is also introduced at the same time. A 1D representation of this process is illustrated in Fig. 1, where a polymer layer in a polymer block is replaced with metal with the same thickness. The total resistance of the new composite is determined by the competition of the high thermal conductivity of the replaced layer and the introduced TBR. Defining the resistance introduced by the metal middle layer as \( R_{\text{intro}} = R_{\text{metal}} + 2R_B \), the net resistance change after the replacement in Fig. 1 can be expressed as

\[
\Delta R = R_{\text{intro}} - R_{\text{poly}}
\]

Here, \( R_B \) is the TBR which is related to the material properties such as lattice mismatch, etc. \( R_{\text{metal}} \) and \( R_{\text{poly}} \) refer to the conduction resistance of metal and polymer layers, respectively, which depend on the middle layer thickness and their thermal conductivities. Therefore, \( \Delta R \) also varies with these factors. In order to obtain a quantitative analysis of the relationship between \( \Delta R \) and the middle layer thickness, expressions for the resistances in Eq. (1) need to be derived.

A. A general solution to two-temperature model equations for a metal thin film sandwiched between two dielectrics

The interfaces in Fig. 1 are the metal/nonmetal interfaces. In metals, thermal transport involves both phonons and electrons. We apply the two-temperature model for analysis, where electrons and phonons are depicted as two interacting subsystems with their own temperatures. Their interaction strength is described by the coupling factor \( G_{ep} \). If we ignore all the external factors such as laser heating, etc., the steady-state governing equations are

\[
\begin{align*}
  k_e \frac{\partial^2 T_e}{\partial x^2} - G_{ep}(T_e - T_p) &= 0, \\
  k_p \frac{\partial^2 T_p}{\partial x^2} + G_{ep}(T_e - T_p) &= 0.
\end{align*}
\]

Here, \( T \) and \( k \) denote the temperature and thermal conductivity, respectively, and \( e \) and \( p \) are the index for electrons and phonons, respectively. In dielectric materials, usually the electrons’ effect can be neglected compared with phonons’ effect. At the interface, the cross-interface electron-phonon interaction may also be ignored if the temperature difference is not large (<1000 K). Based on the above assumptions, only phonons can transfer energy across the interface, while electrons cannot. In a polymer/metal/polymer sandwich system as illustrated in Fig. 2, the boundary conditions are
where \( J \) is the heat flux and \( k_{\text{poly}} \) is the thermal conductivity of the polymer. Different boundary conditions cause nonequilibrium between the two energy carriers in the metal. As illustrated in Fig. 2, electrons and phonons have strong nonequilibrium near the interface where their temperatures deviate from each other. This will render an extra interfacial resistance in addition to the phonon-phonon coupling resistance. As a result, the total TBR consists of two parts: \( R_B = R_{pp} + R_{ep} \).

\( R_{pp} \) can be acquired by several approaches such as the acoustic mismatch model (AMM), diffuse mismatch model (DMM), and MD simulations.\(^{23-25}\) It can be treated as a relatively constant value at a specific temperature. Therefore, only \( R_{ep} \) needs to be derived. Combining Eqs. (2) and (3), we can obtain an expression for the temperature profile in the metal

\[
\begin{align*}
T_e &= T_{mid} - \frac{J}{k_p + k_e} x + \frac{k_p}{k_p + k_e} J_d \text{sinh} \left( \frac{L}{2d} \right) \cdot 2 \cdot \text{sinh} \left( \frac{x}{d} \right), \\
T_p &= T_{mid} - \frac{J}{k_p + k_e} x - \frac{k_e}{k_p + k_e} J_d \text{sinh} \left( \frac{L}{2d} \right) \cdot 2 \cdot \text{sinh} \left( \frac{x}{d} \right).
\end{align*}
\]

Equation (7) is generally applicable regardless of the metal film thickness. If we apply the infinite large-system limit, \( R_{ep} \) recovers the expression in Wang’s work.\(^{20}\)

**B. The critical thickness \( L_c \)**

With expressions for all the resistances in Eq. (1), \( \Delta R \) can be expressed as

\[
\Delta R = \frac{2}{(G_{ep}k_p)^2} \left( \frac{k_e}{k_e + k_p} \right)^2 \text{tanh} \left( \frac{L}{2d} \right) + \frac{2}{h_{pp}} + \frac{L}{k_e + k_p} - \frac{L}{k_{poly}},
\]

where \( h_{pp} \) is the phonon thermal boundary conductance (TBC) which is the inverse of \( R_{pp} \).

If we want to improve the heat conduction in the new composite material, \( \Delta R \) must be negative so that the total resistance decreases. Observing the first-order derivative of \( \Delta R \), it can be shown that

\[
\frac{d\Delta R}{dL} \leq \frac{2}{(G_{ep}k_p)^2} \left( \frac{k_e}{k_e + k_p} \right)^2 - \frac{1}{k_e + k_p} - \frac{1}{k_{poly}}.
\]

For most metals, the value of \( k_e + k_p \) is usually greater than 100 W/m K, and \( G_{ep} \) is on the order of \( 1 \times 10^{16} \) W/m³ K, while \( k_{poly} \) for most common polymer materials is smaller than 10 W/m K. As a result, the right side of Eq. (9) is always negative, which means that \( \Delta R \) decreases monotonically as \( L \) increases, from its maximum value of \( 2/h_{pp} \) at \( L = 0 \). The critical thickness \( L_c \) is defined as the value where \( \Delta R \) crosses zero, and \( L \) has to be larger than \( L_c \) to result in a negative \( \Delta R \). Therefore

\[
\frac{2}{(G_{ep}k_p)^2} \left( \frac{k_e}{k_e + k_p} \right)^2 \text{tanh} \left( \frac{L_c}{2d} \right) + \frac{2}{h_{pp}} + \frac{L_c}{k_e + k_p} - \frac{L_c}{k_{poly}} = 0.
\]

It is noteworthy that \( L_c \) only exists when \( k_{poly} < k_e + k_p \), which is true for most materials. The first term in Eq. (10) makes it difficult to get an explicit analytical solution for \( L_c \). However, we can simplify the equation by taking two extreme limits:

1. If \( G_{ep} \) is very small, which is the case where electrons and phonons have very weak coupling

\[
d = \frac{1}{\sqrt{G_{ep} \left( \frac{1}{k_e} + \frac{1}{k_p} \right)}} \to \infty, \quad L_c = \frac{2}{h_{pp}} \to 0, \quad \text{tanh} \left( \frac{L_c}{2d} \right) \to \frac{L_c}{2d}.
\]

The first term in Eq. (10) evolves as

\[
d \to \infty, \quad L_c = \frac{2}{h_{pp}} \to 0, \quad \text{tanh} \left( \frac{L_c}{2d} \right) \to \frac{L_c}{2d}.
\]
\[ \frac{2}{(G_{ep}k_p)^2} \left( \frac{k_e}{k_e + k_p} \right)^{\frac{3}{2}} \tanh \left( \frac{L_c}{2d} \right) = \frac{2d}{k_p(k_e + k_p)} \frac{k_e}{2d} \]
\[ = \frac{k_eL_c}{k_p(k_e + k_p)}. \]  

From Eqs. (10) and (12), \( L_c \) is expressed as
\[ L_c = \frac{2}{\frac{1}{k_{poly}} - \frac{1}{k_p}}. \]  

It is the same result when electrons are not involved. In fact, if \( L_c/d < 1 \), which is usually the case for metals with weak electron-phonon coupling like gold \((L_c/d = 0.5)\), the above expression is still approximately valid, with an error within 10%.

(2) If \( G_{ep} \) is very large, which is the case that electrons and phonons have very strong coupling
\[ d = \frac{1}{\sqrt{G_{ep} \left( \frac{1}{k_e} + \frac{1}{k_p} \right)}} \to 0, \frac{L_c}{2d} \to \infty, \tanh \left( \frac{L_c}{2d} \right) \to 1. \]  

The first term in Eq. (10) evolves as
\[ \frac{2}{(G_{ep}k_p)^2} \left( \frac{k_e}{k_e + k_p} \right)^{\frac{3}{2}} \tanh \left( \frac{L_c}{2d} \right) = \frac{2d}{k_p(k_e + k_p)} \frac{k_e}{2d} = 0. \]  

Then
\[ L_c = \frac{2}{\frac{1}{k_{poly}} - \frac{1}{k_p} + r_{ep}}, r_{ep} = \frac{k_e}{k_p}. \]  

This is the case when electrons and phonons are in perfect equilibrium, indicating that electrons fully contribute to thermal transport. In fact, if \( L_c/d > 3 \), which is usually the case for metals with strong electron-phonon coupling like nickel \((L_c/d = 4.1)\), the above expression is still approximately valid, with an error of 10%.

(3) When \( L_c/d \) is between 1 and 3, the above two simplifications can no longer give accurate results, and we will need to numerically solve Eq. (10) to get an accurate solution. For a specific pair of materials, one can choose one from the above equations with \( h_{pp}, k_p, \) and \( k_{poly} \) to estimate \( L_c \) first, and then check if \( L_c/d \) falls in the corresponding range. If so then the approximation is valid, otherwise a more accurate calculation is required.

### III. SAM/Au/SAM CASE STUDY

In this section, we will present a case study of a gold thin film sandwiched between aligned SAM chains. The Au/SAM interface has been investigated by several recent studies which could provide benchmarks for our calculations.\(^{28–30}\)

A TTM-MD simulation is performed first to predict the thermal properties of the SAM molecule chain ranges from 0.9 W/m K to 2.4 W/m K, which also agree with previous literature studies.\(^{34,35}\) It is noteworthy that this value is a little bit higher than that of a common polymer matrix. The reason is that these SAM

![Graph showing temperature profile of the Au/SAM/Au/SAM/Au multiplayer system zoomed at the middle layer from the TTM-MD simulation.](Image)
molecule chains in the system are well aligned. The neat structure ensures that the phonon propagation is smooth and results in a reasonably high thermal conductivity.

B. TTM-Fourier calculations

We have developed equations for estimating $L_c$ in Sec. II. However, usually a more accurate result without approximations is desired. Then, we need to go back to Eq. (10) and acquire the solution through numerical methods. The detailed parameters used for the Au/SAM/Au calculation are listed in Table I. Based on the TTM-MD simulation results, we assign $h_{pp}$ as 350 W/m$^2$ K for convenience. For the thermal conductivity of polymer, we use our own averaged value of 1.48 W/m K, which is reasonable compared with Chen’s result of 2.081 W/m K.\textsuperscript{35} $k_e = 313$ W/m K is the bulk electronic thermal conductivity of gold. $G_{ep} = 2.8 \times 10^{16}$ W/m$^3$ K is a commonly used value for gold.\textsuperscript{36} The trend of $\Delta R$ vs. metal film thickness is illustrated in Fig. 4. $\Delta R$ decreases monotonically with the film thickness. And when $\Delta R = 0$, which means that the systems resistance is unchanged, we obtain the critical thickness $L_c = 10.8$ nm. By comparing with the result $L_c = 10.96$ nm given by Eq. (13), we can see that Eq. (13) provides an excellent approximation for metals with weak electron-phonon coupling like gold.

The TTM-Fourier method’s prediction of the temperature profile of the entire system is illustrated in Fig. 5. The metal film thickness is chosen as the critical thickness for the SAM/Au/SAM system. The electrons and phonons are in nonequilibrium except at the middle point. Therefore, it justifies the necessity to use our more general Eq. (7) for $R_{ep}$. The fitted temperature is almost identical to electrons’ temperature, which has a very flat profile, indicating a small electronic contribution to the overall thermal conduction. This agrees with Li’s result which shows that in very thin metal films phonons dominate the energy transport process.\textsuperscript{37}

A closer observation of $R_{intro}$ vs. $L$ also reveals the fact that electrons hardly contribute to the thermal conduction in a thin gold film. As illustrated in Fig. 6, at a thickness smaller than 20 nm $R_{intro}$ mainly comes from $R_{pp}$, while its increasing trend is mainly determined by $R_{ep}$’s increase with $L$. The gradient of $R_{intro}$ is very close to $1/k_p$. This indicates that the $R_{intro}$ curve is almost identical to the straight line with the expression of $R(L) = 2R_{pp} + L/k_p$, which means that the effective thermal transport process is the same as if there is

\begin{table}[h]
\centering
\caption{Thermal properties used in TTM-Fourier calculation.}
\begin{tabular}{ll}
\hline
Thermal property & Value \\
\hline
$h_{pp}$ & 350 MW/m$^2$ K \\
$k_{polymer}$ & 1.48 W/m K \\
k_e & 313 W/m K \\
$k_p$ & 6.41 W/m K \\
$G_{ep}$ & $2.8 \times 10^{16}$ W/m$^3$ K \\
\hline
\end{tabular}
\end{table}
only phonon participation. This stimulates our proposal for a new analytical approach for thermal conduction in sandwiched metal thin films, which will be discussed in Sec. IV A.

IV. DISCUSSION

A. The effective thermal conductivity of the sandwiched metal layer

So far, we have assumed that the thermal conductivity of the metal layer is still \( k_e + k_p \), while we lump \( R_{ep} \) into the interface resistance. This is commonly done for a single interface between semi-infinite metal and dielectric, since electrons and phonons are in equilibrium in nearly the entire metal except for the short cooling region near the interface. However, it is not so intuitive for the sandwiched thin film here since electrons and phonons are in strong non-equilibrium except for the mid-plane, and the thermal conduction is dominated by phonons. Therefore, it is more intuitive to start the metal layer as a phonon-only system, and investigate what effects the electrons will bring. Therefore, here we present an alternative analysis where we define a new effective thermal conductivity of the metal layer while not assuming it to be \( k_e + k_p \) anymore.

From Eq. (7), we can express \( R_{intra} \) as follows:

\[
R_{intra} = 2R_{ep} + R_{metal,c} + 2R_{pp} = \frac{2}{(G_{ep}k_p)^{\frac{1}{2}}} \left( \frac{k_e}{k_e + k_p} \right)^{\frac{1}{2}} \tanh\left( \frac{L}{2d} \right) + \frac{L}{k_e + k_p} + 2R_{pp},
\]

(17)

Previously, we have treated \( R_{ep} \) as part of \( R_B \). Here, we view the metal layer primarily as a phonon system, so we only include \( R_{pp} \) in \( R_B \), while lumping \( R_{ep} \) into the conduction resistance of the metal layer \( R_{eff} \). Therefore,

\[
R_{eff} = 2R_{ep} + R_{metal} = \frac{2}{(G_{ep}k_p)^{\frac{1}{2}}} \left( \frac{k_e}{k_e + k_p} \right)^{\frac{1}{2}} \tanh\left( \frac{L}{2d} \right) + \frac{L}{k_e + k_p},
\]

(18)

and then define the effective thermal conductivity of the metal film as

\[
k_{eff} = \frac{R_{eff}}{L} = \frac{k_e + k_p}{1 + \frac{2k_e}{L} \left[ k_e \left( \frac{k_e}{G_{ep}k_p(k_e + k_p)} \right)^{\frac{1}{2}} \tanh\left( \frac{L}{2d} \right) \right]^{-1}}.
\]

(19)

The corresponding thermal circuit is shown in Fig. 7. And we can easily find that

\[
k_{eff} \to k_p, \quad \text{when } L \to 0 \text{ or } G_{ep} \to 0,
\]

\[
k_{eff} \to k_e + k_p, \quad \text{when } L \to \infty \text{ or } G_{ep} \to \infty.
\]

(20)

This indicates that: (1) in a sandwiched metal film that is very thin (or has very weak electron-phonon coupling), the effective thermal conductivity is identical to its lattice portion and electrons are as if not involved at all. We call this the “thin limit.” And (2) when the metal is thick (or has strong electron-phonon coupling), the effective thermal conductivity recovers the bulk value, and we call it the “thick limit.”

We expect that this interpretation, especially the thin limit, to be very useful for experimentalists. In experiments, \( R_{pp} \) is often treated as the only TBR. It is then straightforward to match the measured values with components of the thermal circuit in Fig. 7. If the thin limit applies, \( k_{eff} \) should become the lattice thermal conductivity while not the bulk metal conductivity \( k_e + k_p \). This provides an intuitive way to check and interpret experiments.

A more detailed dependence of \( k_{eff} \) on the film thickness is illustrated in Fig. 8. Gold is still taken as the example here. It can be observed that the thin limit is valid when the thickness is much smaller than the electron-phonon cooling length of 45 nm, as \( k_{eff} \) remains below 110% of \( k_p \) when \( L \) is below 16.2 nm. In this range, which is usually the case for a metal thin film sandwiched between dielectric materials, our new approach where \( R_{ep} \) is included as part of the film conduction resistance is more intuitive. Our model bypasses electrons’ effect and only requires lattice temperature measurements for analysis, and the measured effective thermal conductivity \( k_{eff} \) is simply \( k_p \). As \( L \) increases, \( k_{eff} \) starts to deviate from \( k_p \) and eventually converges to \( k_e + k_p \). For a thickness larger than 10.2 μm, which is more than 200 times larger than the cooling length, \( k_{eff} \) can be approximated as \( k_e + k_p \) with an error smaller than 10%. In this range, which is usually the case when investigating an interface between two semi-infinite blocks, the original thermal circuit where \( R_{ep} \) is part of the TBR works better. The experimentally measured \( k_{eff} \) can no longer be interpreted by \( k_p \). When \( L \) falls in the transition region marked in Fig. 8, an accurate analysis requires utilization of the exact form of Eq. (18) and investigation of the corresponding serial thermal circuit.

B. Factors affecting \( L_c \)

In this section, we discuss several factors that can affect \( L_c \) based on Eq. (10): (1) \( R_{pp} \), (2) \( G_{ep} \), (3) \( k_p \), (4) \( k_{poly} \), and (5) \( R_{ep} \).

(1) \( R_{pp} \) comprises the most part of TBR when the metal film is as thin as ~10 nm. A larger \( R_{pp} \) will increase TBR, thus increasing the critical thickness. For example, if one uses the experimentally determined \( R_{pp} \) which is much higher than the one we used in our previous
calculations, the resulting \( L_c \) will be larger than 10.8 nm, but the general trend is the same and our conclusions are still valid.

(2) \( G_{ep} \) measures the electron-phonon coupling strength and is directly related to \( R_{ep} \). A larger \( G_{ep} \) will render a smaller \( R_{ep} \) and thus a smaller TBR, resulting in a smaller critical thickness.

(3) \( k_p \) dominates the thermal transport process in thin films as mentioned above. As a result, it is almost identical to the effective thermal conductivity \( k_{eff} \). When \( k_p \) increases, \( k_{eff} \) increases, \( R_{ep} \) will decrease according to Eq. (7); hence, the critical thickness will be smaller.

(4) \( k_{poly} \) is the thermal conductivity of the polymer matrix. A larger \( k_{poly} \) will increase the difficulty for the metal film to compensate for the resistance increase due to introduced TBR after the replacement. Therefore, larger \( k_{poly} \) will result in larger \( L_c \).

(5) \( r_{ep} \), which is the ratio of \( k_e \) over \( k_p \), can affect \( R_{ep} \) as well as the thermal conductivity of the metal film. If we keep \( k_p \) as fixed, larger \( r_{ep} \) will increase \( k_e + k_p \) and also change \( R_{ep} \) (whether it will increase or decrease depends). Generally, a larger \( k_e \) is beneficial and will result in a smaller \( L_c \). The effect is more significant if the values of \( k_e \) and \( k_p \) are comparable which means that \( r_{ep} \) is around 1, especially for large \( G_{ep} \).

When we choose the filler material to add into the polymer matrix, two characteristic properties are \( G_{ep} \) and \( k_e \). Here, we plot \( G_{ep} \) and \( k_e \)'s effect on \( L_c \) in Fig. 9 on an arbitrary SAM/Au/SAM system to gain a more straight-forward insight. \( k_p \) is fixed at the value of gold’s lattice thermal conductivity, and \( k_e \) is represented by the normalized parameter \( r_{ep} \). \( L_c \) decreases with larger \( G_{ep} \) and larger \( k_e \). However, \( k_e \)'s effect is only significant when \( k_e \) is small and the impact vanishes as \( k_e \) increases. However, metals usually have much larger \( k_e \) than \( k_p \), so \( r_{ep} \) is not so significant in controlling \( L_c \).

V. SUMMARY

We have proposed a TTM-Fourier approach with input parameters from TTM-MD to predict the critical thickness \( L_c \) for metal particle-polymer composite in 1D. A general solution to TTM for thermal transport in a metal film sandwiched between dielectric materials is derived. As an example, \( L_c \) for the 1D SAM/Au/SAM system is calculated to be around 10.8 nm. Based on the theoretical equations, we define an effective thermal conductivity for sandwiched metal thin films and propose a new thermal circuit analysis that are intuitive to interpret experimental results. A detailed discussion on the applicable range of our model is presented. It is shown that when the metal film is much thinner than its electron-phonon cooling length (thin limit), the effective thermal conductivity reduces to just the phonon part. For a metal layer with a large thickness (thick limit), the conventional thermal circuit is more advantageous, and the effective thermal conductivity recovers the metal bulk value. For the thickness in between the two limits, our general TTM solution still provides an accurate method for analysis. Finally, several factors affecting \( L_c \) are discussed, and it is discovered...
that the thermal conductivity, TBR, and the electron-phonon coupling factor all play important roles in determining $L_c$, which can provide us with a general guidance in the choice of materials when synchronizing a new composite.