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Nanoscale Heat Conduction across Metal-Dielectric Interfaces

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ABSTRACT

We report a theoretical study of nanoscale heat conduction across nanolaminates consisting of alternating layers of metal and dielectric materials. Nanolaminates are promising as thermal barrier coatings for energy generation and conversion applications because they offer unique opportunities to achieve superior thermal performance without compromising mechanical strength or chemical protection characteristics. A continuum two-fluid energy transport equation is solved to predict the thermal resistance of a metallic film bounded by dielectric materials. Analysis of existing experimental data is consistent with the present model, suggesting that electron-phonon spatial non-equilibrium plays an important role in heat conduction across metal-dielectric interfaces.

Heat conduction across interfaces [1] is a critical consideration in nanolaminates that consist of alternating layers of disparate materials. This is because the thermal interface resistance often dominates the total thermal resistance due to high density of interfaces present [2-4]. Nanolaminates are promising as thermal barrier coatings for energy generation and conversion applications because they offer unique opportunities to achieve superior thermal performance without compromising mechanical strength or chemical protection characteristics.

Theoretical and experimental studies have been conducted in the past to gain fundamental understanding of thermal resistance at interfaces between dissimilar solids [1,5].

Detailed quantitative understanding of the thermal boundary resistance, however, is still incomplete. In particular, previous experimental studies often reported values that are larger than theoretical predictions near room temperature and above. Better agreement has been observed at low temperatures, typically below 50K, for interfaces where mismatch in lattice vibrational properties dominates thermal boundary resistance.

Various mechanisms have been proposed to explain discrepancy between theoretical predictions and experimental data. The presence of defects or contamination layers has most often been cited as possible mechanisms. For interfaces involving crystalline dielectric substrates, surface polishing procedures can create damaged layers. Structural disorder in the damaged layers can strongly impede heat conduction, which results in extra thermal resistance across the interface. Previous studies also observed that details of surface cleaning procedure can affect thermal boundary resistance.

The author suggested earlier that inefficient coupling between electrons and phonons is responsible for large thermal resistance at metal-dielectric interfaces. In other words, abrupt change in the dominant heat carriers across the interface introduces extra thermal resistance. Analogous non-equilibrium effects were considered in previous studies of energy transport across superconductor-dielectric interfaces [6] and across interfaces between TiN films and MgO single crystal substrates.

The present manuscript examines the impact of non-equilibrium between electrons and phonons on the thermal resistance of laminates consisting of alternating layers of metal and amorphous dielectric films. A continuum two-fluid energy transport equation is solved to predict the thermal resistance of a metallic film bounded by dielectric materials. Analysis of existing experimental data is consistent with the present model, suggesting that electron-phonon spatial non-equilibrium plays an important role in heat conduction across metal-dielectric interfaces.

Previous studies of the thermal interface resistance between a metal and a dielectric material assumed that electrons and phonons within the metal are in equilibrium with each other. Under this assumption the out-of-plane thermal resistance of the metal is very often negligible due to its large electronic thermal conductivity. The validity of this assumption, however, needs more careful attention. In the absence of significant inelastic scattering at the interface, electrons in a metal interact indirectly with atomic vibrations in adjacent dielectric materials. Electrons first exchange energy with phonons within the

metal, which are then coupled with atomic vibrations in the dielectric materials through interface bonds. As a result of such selective coupling, electron and phonon temperatures at the interface deviate from each other. The impact of such non-equilibrium is especially important for metals with weak electron-phonon coupling, such as Au and other noble metals.

The present work solves the coupled energy conservation equations for phonons and electrons in metal films based on the continuum two-fluid model:

$$-\frac{d}{dx} \left(k_e \frac{dT_e}{dx} \right) = -G(T_e - T_{ph}) + q'' \quad (1)$$

$$-\frac{d}{dx} \left(k_{ph} \frac{dT_{ph}}{dx} \right) = G(T_e - T_{ph}) \quad (2)$$

Electrons and phonons are each assumed to be in separate equilibrium states as characterized by the two temperatures T_e and T_{ph} . Subscripts e and ph are used to label quantities relevant to electrons and phonons, respectively. The electron-phonon coupling constant is denoted by G and the thermal conductivities by k . The two-fluid model has widely been used in the studies of ultra-short pulse laser heating of metals [8]. We emphasize, however, that the present work employs the model to capture spatial as opposed to temporal non-equilibrium in electron-phonon systems. While such a non-equilibrium process can be studied more rigorously using the Boltzmann transport equation, it involves much greater mathematical complexity and does not readily yield physical insights or useful analytic expressions.

We restrict ourselves to a metal film of thickness L that is sandwiched between two amorphous dielectric materials. Since the mean free path of heat carriers in amorphous insulators is of the order of interatomic spacing, heat conduction can be described using the continuum model down to nanoscales. The boundary conditions are:

$$T_{ph}|_{x=0} = T_0, \quad T_{ph}|_{x=L} = T_L, \quad \left. \frac{dT_e}{dx} \right|_{x=0,L} = 0. \quad (3)$$

The third condition reflects that energy transport by electrons is confined within the metal film. Analytic solutions to Eqs. (1-3) can be obtained if all the material parameters are assumed to be constant. The solutions neglect the size effect on the properties but can serve as a first-order approximation for nano-scale thin films that are highly defective and consist of nanoscale grains. We can evaluate the thermal resistance of the metal film as

$$R''_{metal} = \frac{T_{ph}|_{x=0} - T_{ph}|_{x=L}}{q''} = \frac{L}{k_{ph} + k_e} + 2 \left(\frac{k_e}{k_{ph}} \right) \left(\frac{\delta}{k_{ph} + k} \right) \left(\frac{e^{L/\delta} - 1}{e^{L/\delta} + 1} \right), \quad (4)$$

The first term, proportional to film thickness L , is identical to the solution of the conventional heat diffusion equation. The second term, to be labeled R''_{NE} , arises from spatial non-equilibrium between phonons and electrons near the interface. The relative impact of R''_{NE} is small at low temperatures because the intrinsic boundary resistance R''_b increases rapidly with decreasing temperature. The intrinsic boundary resistance is associated for example with mismatch in atomic vibrational properties. Since the

magnitude of R''_{NE} depends on metal thickness, the effective interface resistance, which is the sum of R''_{NE} and R''_B , is also a function of metal film thickness.

An important parameter identified in Eq. (4) is the electron-phonon coupling distance δ .

The parameter is defined as

$$\delta = \left(\frac{k_e k_{ph}}{G(k_e + k_{ph})} \right)^{1/2}. \quad (5)$$

It can be interpreted as the characteristic length of a region near the interface where electrons and phonons remain out of equilibrium with each other. Heat conduction across this region is mostly by phonons alone.

The electron-phonon coupling constant G can be determined by analyzing ultra-short pulsed laser heating experiments [9,10]. The coupling constant was found to be small for noble metals ($G_{Au} \sim 3 \times 10^{16}$ W/m³ K) and large for metals in the trivalent main group and the chrome congener family ($G_{Al} \sim 25 \times 10^{16}$ W/m³ K, $G_{Cr} \sim 10 \times 10^{16}$ W / m³ K). The electronic thermal conductivity k_e can be estimated from the electrical conductivity using the Wiedemann-Franz law. The phonon thermal conductivity of metals near room temperature and above is believed to be influenced primarily by phonon-phonon Umklapp scattering [11]. Estimates of the phonon thermal conductivity can be made from theoretical models using the atomic mass, Debye temperature, and the Gruneisen constant as input parameters [12]. In bulk samples of pure copper the lattice conductivity is estimated to be 5 W/m K at room temperature, accounting for only about 1% of the

total thermal conductivity. This result is consistent with a value extrapolated from the analysis of experimental data on Cu alloys where electronic contribution can be predicted with better confidence. Based on such estimates, we anticipate that the values of δ for typical metals are of the order of a few nanometers near room temperature.

To gain more insight, we examine the two limiting cases. In the limit $L \gg \delta$, one can approximate Eq. (4) as:

$$(R''_{metal})_{thick} \approx L/(k_e + k_{ph}) + 2\delta/k_{ph}. \quad (6)$$

It is assumed that electrons dominate heat transport ($k_e \gg k_{ph}$) within the metal. The non-equilibrium component is equivalent to the thermal resistance that would result if the energy is transported only by phonons over the distance δ . In the opposite limit ($L < \delta$), the thermal resistance increases approximately linearly with film thickness:

$$(R''_{metal})_{thin} \approx L/k_{ph}. \quad (7)$$

Note that the thermal conductivity in the denominator of Eq. (7) is the phonon thermal conductivity and not the sum of the electronic and phonon thermal conductivity. In this limit electrons and phonons are decoupled from each other and only phonon mediated heat conduction is significant.

Figure 1 shows existing experimental data on the thermal resistance per unit thickness of nano-laminates consisting of alternating W and amorphous AlOx layers [3]. The dashed line corresponds to values predicted by the conventional heat conduction equation with a constant thermal interface resistance. The thermal conductivity of AlOx films was determined experimentally. The Wiedemann-Franz law was used together with the measured in-plane electrical conductivity to estimate the electronic thermal conductivity of W films.

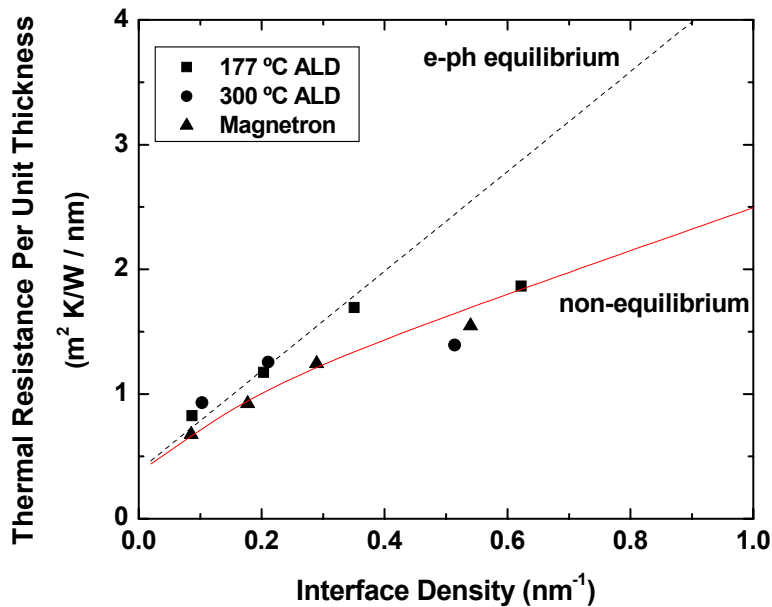


Figure 1: Predicted and measured thermal resistance across W-AlOx nano-laminates.

The data are from ref 3. The dotted line is the prediction of the conventional heat conduction equation. The solid line is the prediction of the two-fluid energy transport model that takes into account spatial electron-phonon non-equilibrium.

The thickness-independent interface resistance is obtained from a fit to the data from samples with interface density below 0.2 nm^{-1} . The prediction of the conventional heat conduction model is consistent with the data for laminates with thick constituent layers is considerably larger than the data for nano-laminates with thinner constituent layers.

We next apply the non-equilibrium thermal resistance model described earlier to analyze the same set of data. Equation (4) is used to evaluate the thermal resistance of the metal layer. Our goal is not to prove the absolute validity of the present model but to examine whether a physically reasonable set of parameters can describe the experimentally observed behavior. The solid line in Fig. 1 corresponds to a prediction of the present model using the parameters obtained from a fit to the data: $k_{ph} = 1 \text{ W/m K}$, $R_B'' = 1.7 \times 10^{-9} \text{ m}^2 \text{ K/W}$, $\delta = 2 \text{ nm}$. While no rigorous theoretical models/material characterization data are available to accurately predict each of these parameters, they are all within physically reasonable limits. This suggests that spatial phonon-electron non-equilibrium does play a role in the thermal resistance of nano-laminates. The fit value for the phonon thermal conductivity is close to the minimum thermal conductivity value predicted using material parameters of fully dense crystalline materials. This is not unreasonable since the actual films are expected to be less dense and defective. Only a very crude estimate can be made for the thermal boundary resistance R_B'' . The diffuse mismatch theory under the Debye approximation predicts a thermal boundary resistance of $3 \times 10^{-9} \text{ m}^2 \text{ K/W}$, which compares favorably with the fit value. The extracted electron-phonon coupling constant is $22 \times 10^{16} \text{ W/m}^3 \text{ K}$, comparable to that of Al and Cr.

While the present study identifies one potential mechanism responsible for apparent thickness dependence of metal-dielectric interface resistance, other mechanisms may have played roles. One such mechanism is heat conduction by electrons through pin holes in the dielectric layers. To explain the data using this mechanism, however, the electrical conductance across the nano-laminates must be significantly higher than that of tunnel junctions that incorporate even thinner dielectric layers. Simultaneous electrical and thermal characterization of nano-laminates is necessary to rule out this mechanism.

Previous studies attributed high thermal resistance at interfaces between metal and crystalline dielectric materials to the presence of defects or highly disordered layers near the interface [1]. Other theoretical studies investigated possible impact of phonon dispersion, either by modifying the acoustic or diffuse mismatch model or by performing lattice dynamics calculations [5]. For amorphous dielectric films, however, we do not expect drastic variations in structures, defect density, or vibrational spectra with thickness.

The excitation of interface modes may also lead to decrease in the thermal interface resistance. Such decrease was predicted to be significant for interfaces between materials with very different Debye temperatures, such as Pb films on diamond [13]. Transmission of evanescent waves, or phonon tunneling, may be important in nano-laminates. Detailed theoretical studies, most desirably first-principle based calculations, are needed to further examine these mechanisms.

In summary, the present work applies the two-fluid continuum energy transport model to investigate the impact of non-equilibrium between electrons and phonons on heat conduction across nano-laminates. An analytic expression for the thermal resistance of a metal layer sandwiched between dielectric materials is derived as a function of its thickness. The present model is consistent with the existing experimental data on the thermal resistance of W/AlOx nano-laminates, suggesting that spatial non-equilibrium between electrons and phonons plays an important role in energy transport across alternating layers of metallic and insulating materials. The present work contributes to improving fundamental understanding of nanoscale energy transport to help guide the design of superior thermal barriers based on nano-laminates.

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