

Phonon Dominated Heat Conduction Normal to Mo/Si Multilayers with Period below 10 nm

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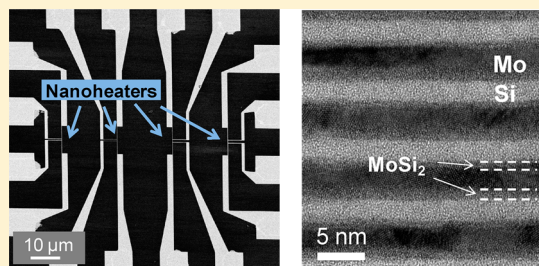
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S Supporting Information

ABSTRACT: Thermal conduction in periodic multilayer composites can be strongly influenced by nonequilibrium electron–phonon scattering for periods shorter than the relevant free paths. Here we argue that two additional mechanisms—quasiballistic phonon transport normal to the metal film and inelastic electron–interface scattering—can also impact conduction in metal/dielectric multilayers with a period below 10 nm. Measurements use the 3ω method with six different bridge widths down to 50 nm to extract the in- and cross-plane effective conductivities of Mo/Si (2.8 nm/4.1 nm) multilayers, yielding 15.4 and 1.2 W/mK, respectively. The cross-plane thermal resistance is lower than can be predicted considering volume and interface scattering but is consistent with a new model built around a film-normal length scale for phonon–electron energy conversion in the metal. We introduce a criterion for the transition from electron to phonon dominated heat conduction in metal films bounded by dielectrics.

KEYWORDS: Nanoscale heat conduction, phonon scattering, metal–dielectric interface, Mo/Si multilayers, thermal interface resistance



Heat conduction across metal interfaces with dielectrics and semiconductors governs the behavior of many nanostructured materials and devices. Past research observed the importance of interface resistance in metal/dielectric multilayers with periods of a few nanometers, such as W/ Al_2O_3 thermal barrier coating¹ and Ta/ TaO_x tunnel junctions.² Conduction normal to these interfaces is complicated by thermal energy conversion between electrons and phonons near and at the interface. This conversion facilitates the lowest possible thermal resistance because phonons are the dominant heat carriers in dielectrics and most semiconductors, while electrons are the dominant heat carriers in metals. Extensive research has been performed on the metal–dielectric interface thermal resistances for the situation where the metal region is thick compared to the carrier free paths, which justifies the assumption that the energy conversion is complete and the dominant carrier types on each side are fully responsible for conduction.^{3–7} However, there are a variety of applications in which the small thickness of the metal film inhibits the electron–phonon coupling and, we argue in this paper, can render phonons the dominant heat conductor in the direction normal to the metal film. One example is Mo/Si multilayers with an ~ 7 nm period, which serve as mirrors in extreme ultraviolet (EUV) and soft X-ray optics.⁸ Heat conduction in these mirrors can strongly influence performance and reliability due to thermally induced stress and atomic diffusion between Mo and Si thin films.^{9,10}

The present work makes progress on the modeling and experimental investigation of the complex electron and phonon

transport and interaction processes in metal/dielectric multilayers with metal thickness below the electron and phonon mean free paths. We use frequency domain thermometry to measure the in- and cross-plane thermal conductivities of a Mo/Si multilayer sample with 6.9 nm period. Thermal conduction in such multilayers is influenced by phonon–phonon coupling across the interface,³ quasiballistic phonon transport across the metal layer, the electron–phonon nonequilibrium in the metal film,^{4,11,12} and the possibility of significant inelastic electron–interface scattering.⁵ Electron–phonon nonequilibrium has been a common theme for decades in ultrashort pulsed laser heating experiments on metals,^{5,13–15} but the potential for strong steady-state nonequilibrium has received relatively little attention. In this work we show that ultrathin metal films spatially amplify the relevance of near-interfacial nonequilibrium in the steady state, which suppresses the electron contribution to conduction in the film-normal direction and renders phonons the dominant heat carriers.

Multilayers consisting of 40 Mo/Si bilayers with a period thickness of 6.9 nm are deposited by dc magnetron sputtering. The ratios of the Mo layer thickness to the bilayer period studied here include $\Gamma = 0.4$ and 0.6. These values are selected based on the practical requirement of optimal reflectivity in the EUV mirror applications.^{8,9,16} Preliminary sheet resistance

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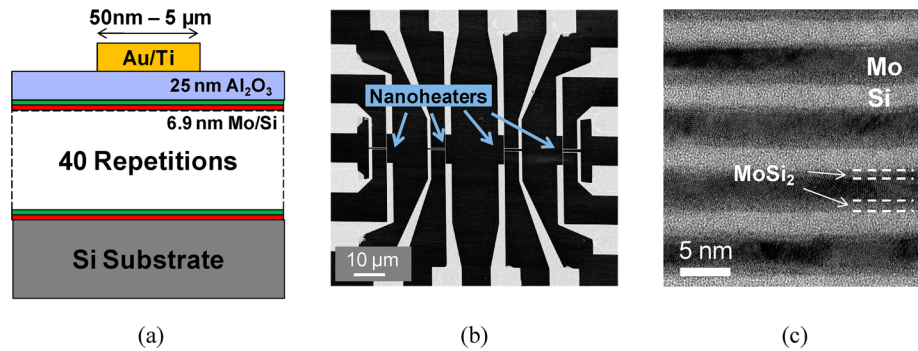


Figure 1. Mo/Si multilayer sample and nanoheater patterns. (a) Schematic of the 40 repetitions of Mo/Si multilayer. (b) Scanning electron microscope (SEM) image of the e-beam patterned Au nanoheaters with width ranging from 50 nm to 5 μm (not all widths are shown in the graph). (c) TEM image of the Mo/Si multilayer. A thin MoSi_2 layer of 0.5–1.2 nm is formed between Mo and Si due to atomic diffusion.

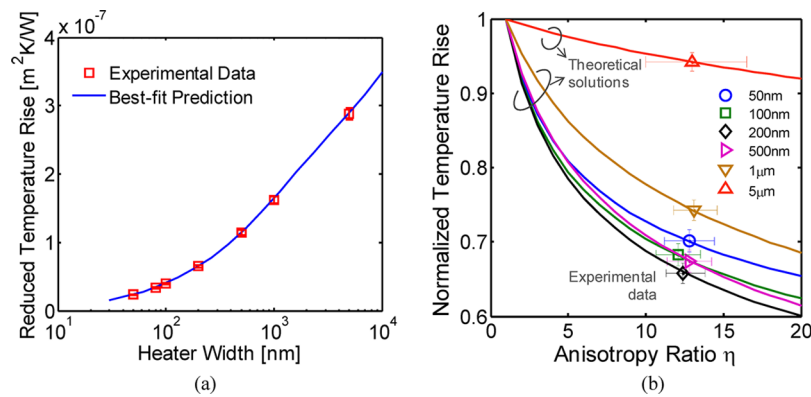


Figure 2. Electrical thermometry data and comparison with best-fit predictions for the Mo/Si multilayer with Mo thickness fraction of $\Gamma = 0.4$. (a) Reduced temperature rise for different heater widths. The numerical fit with all the data points reduces the uncertainty in the fitting of values of k_x and k_z of Mo/Si multilayers as well as the thermal boundary resistance between Al_2O_3 and Mo/Si. (b) Thermal conductivity anisotropy ratio $\eta = k_x/k_z$ is measured with multiple heater widths. The measured anisotropy ratio (~ 13) is consistent among all the heater length scales.

measurements suggest a highly conductive surface of the Mo/Si stack, which motivates the use of a dielectric layer to achieve insulation from the metal heaters on top. An amorphous Al_2O_3 layer is sputtered to provide electrical insulation, as shown in the schematic in Figure 1a. The gold nanoheaters varying in width from 50 nm to 5 μm , as shown in Figure 1b, are patterned by electron beam lithography on the same sample to minimize sample-to-sample variation. A titanium film of 5 nm enhances the adhesion between gold and the insulation layer. The transmission electron microscope (TEM) image in Figure 1c shows the structural detail of the multilayers and thin films of MoSi_2 at the interfaces due to natural atomic diffusion during fabrication. The same fabrication process was utilized to prepare a reference sample with the Al_2O_3 but without Mo/Si multilayers in order to separate the substrate/insulator and Mo/Si contributions to the measured thermal resistance. The Mo/Si multilayers maintain thermal stability at relatively low temperatures (25–100 $^\circ\text{C}$),¹⁷ although interdiffusion between Mo and Si films can become a critical concern at higher temperatures and will be the subject of future thermal conduction studies.⁹

Metal heaters of widths varying from 50 nm to 5 μm extract the in- and cross-plane thermal conductivities of the Mo/Si multilayers by the 3ω technique.^{18,19} A driving ac current at frequency ω generates Joule heating and temperature oscillation in the heater at frequency 2ω . The linear relationship between temperature and the electrical resistivity of gold over the temperature range of this experiment creates a third

harmonic component in the voltage across the heater. This 3ω voltage component is governed in part by the thermal conductivity of the underlying Mo/Si layer. Heaters as wide as 5 μm induce nearly one-dimensional heat conduction through the Mo/Si multilayer and are therefore more sensitive to the cross-plane thermal conductivity, whereas heaters narrower than 200 nm are more suitable for capturing the in-plane heat spreading through the multilayers.

By modeling the 40 repetitions of the Mo/Si multilayers as a single layer with anisotropic thermal conductivity, the average temperature rise ΔT at the surface of a multilayer system can be derived as²⁰

$$\Delta T = \frac{P}{2\pi L b^2} \int_0^\infty \frac{B^+(m) + B^-(m)}{B^-(m)A^+(m) - B^+(m)A^-(m)} \frac{\sin c^2(mb)}{k_{n,z}\gamma_n m^2} dm \quad (1)$$

$$\gamma_n = \sqrt{\eta_n m^2 + i\omega/D_n} \quad (2)$$

where P , L , and b are the heating power, length and width of the heater, respectively. The dimensionless terms $A^+(m)$, $A^-(m)$, $B^+(m)$, and $B^-(m)$ are determined by a recursive matrix procedure developed by J. H. Kim et al.²⁰ The parameters $k_{n,z}$, η_n and D_n are the cross-plane thermal conductivity, anisotropy ratio (k_x/k_z , where x and z denote in- and cross-plane directions, respectively), and thermal

diffusivity of the n^{th} layer, respectively. Numerical solutions of eqs 1 and 2 demonstrate that the temperature rise is most sensitive to the thermal conductivity anisotropy ratio when the heater width is small compared to the film thickness. However, the narrowest heater (50 nm) in this study yields suboptimum sensitivity to the thermal conductivity anisotropy because the heat is largely confined within the 25 nm Al_2O_3 insulation layer before penetrating into the Mo/Si multilayers. In the frequency domain, the 3ω measurement is carried out with an ac driving current between 1 and 20 kHz, a frequency range where the characteristic thermal diffusion depth is larger than the Mo/Si film thickness while still preserving the substrate as a semi-infinite medium.

The measured temperature rises from different heater widths capture the thermal conductivity anisotropy of the Mo/Si multilayer system. Because even the widest heater cannot completely eliminate the lateral heat spreading, simultaneous fitting of all the temperature data obtained from available heater widths from 50 nm to 5 μm provides redundancy and improves the accuracy of the conductivity magnitudes. Figure 2a plots the measured reduced temperature rise, which is the surface temperature rise divided by the heat flux and the theoretical fit for varying heater widths. The thermal conductivity of the Al_2O_3 film is measured independently with a reference sample (25 nm Al_2O_3 on Si substrate) as $k_{\text{Al}_2\text{O}_3} = 3.66 \pm 0.16$ W/mK, which is consistent with previous work^{21,22} and much lower than the bulk crystalline value owing to material disorder. The experimental uncertainty is governed by those of the temperature coefficient of resistivity (TCR) of the gold heaters as well as the parameter variations that are possible in the data fitting process. Solutions of eqs 1 and 2 using the measured Mo/Si anisotropic thermal conductivity data simulate the surface temperature rises, which are normalized to the isotropic case, as shown by the solid curves in Figure 2b. The thermal conductivity anisotropy ratios $\eta = k_x/k_z \approx 13$ are consistent among all the heater widths considering the uncertainty bars. Varying the Mo thickness ratio Γ between 0.4 and 0.6 is expected to have negligible effect on η because the cross-plane thermal conduction is dominated by the interface resistance. Table 1 compiles the measured thermal conductivity data for

Table 1. Effective Cross-Plane Thermal Conductivities of Two Multilayer Samples with Different Mo Thickness Fractions ($\Gamma = d_{\text{Mo}}/d_{\text{total}}$)

Mo thickness fraction Γ	d_{total} (nm)	d_{Mo} (nm)	d_{Si} (nm)	$k_{z,\text{model}}$ (W/mK)	$k_{z,\text{measurement}}$ (W/mK)
0.4	6.9	2.8	4.1	1.30	1.2 ± 0.1
0.6	6.9	4.1	2.8	1.49	1.4 ± 0.1

multilayers with $\Gamma = 0.4$ and 0.6, together with the theoretical calculations from the following sections. The data for the sample with $\Gamma = 0.6$ are taken using the time-domain thermoreflectance method.

The goal of the model developed here is to capture the effects of a variety of complex electron and phonon transport phenomena in thin metal films bounded by dielectrics, especially near and at the metal–dielectric interface. While there are major approximations involved, the purpose is to introduce the dominant physical mechanisms and provide an estimate of their relative importance. Phonon–phonon transmission and scattering at interfaces is estimated here using the diffuse mismatch model (DMM).^{3,23} Electron energy exchange

with phonons within the metal is captured using a two-temperature model (TTM) and a conductance G between the two carrier types.^{4,24,25} Electrons may scatter inelastically at the metal–dielectric interface and interact directly with the phonon system across the interface through an electron–interface conductance.¹³ A consequence of the extra resistances imposed on electron transport normal to the metal films is that phonon heat conduction, usually negligible in metals, can become the dominant mechanism. The following sections investigate the contributions of these mechanisms to the overall cross-plane thermal resistance.

For a heat flux perpendicular to the multilayer surface, the overall thermal resistance consists of both the volumetric thermal resistance of each layer and the thermal boundary resistance at each interface. This approach neglects the thin MoSi_2 layers at the interfaces due to atomic diffusion. The cross-plane thermal resistance of one period of the Mo/Si multilayer, $R_{\text{Mo/Si}}$ is therefore approximated using

$$\begin{aligned}
 R_{\text{Mo/Si}} &= \frac{d_{\text{Mo}} + d_{\text{Si}}}{k_{\text{eff},z}} \\
 &= R_{\text{Mo}} + R_{\text{Si}} + R_{b,\text{Mo-Si}} + R_{b,\text{Si-Mo}} \\
 &= \frac{d_{\text{Mo}}}{k_{\text{Mo},z}} + \frac{d_{\text{Si}}}{k_{\text{Si},z}} + \frac{1}{h_{b,\text{Mo-Si}}} + \frac{1}{h_{b,\text{Si-Mo}}} \quad (3)
 \end{aligned}$$

where d_{Mo} and d_{Si} are the individual thicknesses of the Mo and Si layers, respectively. The effective cross-plane conductivity $k_{\text{eff},z}$ describes the Mo/Si multilayer, while $k_{\text{Mo},z}$ and $k_{\text{Si},z}$ are the cross-plane thermal conductivities of each layer considering the thin film size effects.²⁶ The thermal boundary conductance h_b between a metal and dielectric is related to electron–phonon nonequilibrium and the electron–interface inelastic scattering.

The TTM is traditionally used to describe the electron–phonon nonequilibrium in the metal layer near the metal–semiconductor interface.^{4,11} Electron and phonon temperatures deviate from each other in the near-interface region, often referred to as the cooling length,²⁷ causing electrons to lose energy to phonons which then carry the heat across the interface into the dielectric medium. For steady-state heat flow across the film, the energy balance in the metal can be expressed as

$$\begin{aligned}
 k_e \frac{\partial^2 T_e}{\partial z^2} - G(T_e - T_p) &= 0; \\
 k_p \frac{\partial^2 T_p}{\partial z^2} + G(T_e - T_p) &= 0 \quad (4)
 \end{aligned}$$

where T is temperature, and the subscripts e and p denote electron and phonon, respectively. The electron–phonon coupling coefficient G can be derived from the rate of electron–phonon energy exchange considering an electron density of states (DOS) applicable at room temperature:^{15,28}

$$\begin{aligned}
 G(T_e - T_p) &= \left. \frac{\partial E_e}{\partial t} \right|_{\text{ep}} \\
 &= \frac{4\pi}{\hbar} \sum \hbar \omega_Q |\mathbf{M}_{kk'}|^2 [S(k, k')] \delta(\epsilon_k - \epsilon_{k'} + \hbar \omega_Q) \quad (5)
 \end{aligned}$$

where E_e is electron energy, and ω is the phonon frequency. The electron and phonon wave numbers are denoted as k and Q , respectively, and the electron–phonon scattering matrix $\mathbf{M}_{kk'}$ describes the probability of electron scattering from state k to

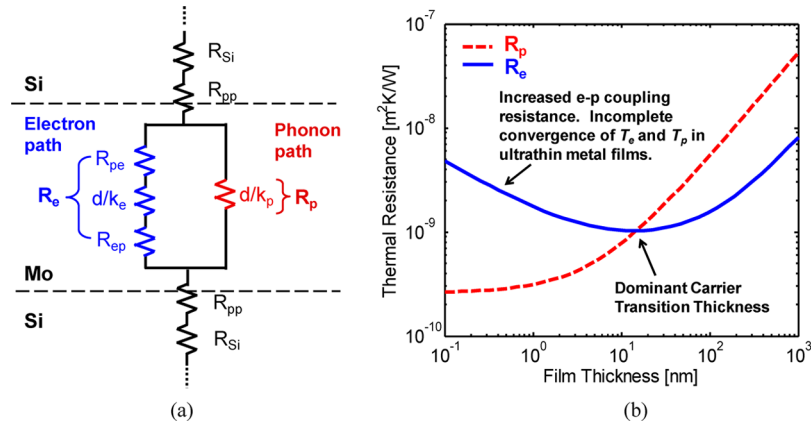


Figure 3. Thermal resistances in the Mo/Si multilayer system. (a) Approximate thermal resistance network model where the conduction in the Mo layer consists of a parallel of the electron path and the phonon direct transmission path. The electron–phonon nonequilibrium resistance, R_{pe} and R_{ep} , result from the conversion of energy between the carrier types and are components of the electron path. (b) Theoretical calculation result shows that the thermal resistance of the phonon path (R_p) becomes smaller than the electron path (R_e) for a metal film thickness of less than $d_{trans} = 14$ nm.

state k' . The thermal statistical factor $S(k, k') = (f_k - f_{k'}) n_Q - f_{k'} (1 - f_k)$ relates the phonon absorption and emission processes to the electron distribution f_k and phonon distribution n_Q . Using the Fermi-Dirac and Bose–Einstein distributions for the electron and phonon systems, respectively, eq 5 can be simplified as

$$\left. \frac{\partial E_e}{\partial t} \right|_{ep} = 2\pi g(\epsilon_F) \int_0^\infty \alpha^2 F(\epsilon, \epsilon', \Omega) (\hbar\Omega)^2 [n(\Omega, T_p) - n(\Omega, T_e)] d\Omega \quad (6)$$

where $g(\epsilon_F)$ is the electron DOS at the Fermi level, and $\alpha^2 F(\epsilon, \epsilon', \Omega)$ is the electron–phonon spectral function.²⁸ Near room temperature, it is reasonable to assume the independence of $\alpha^2 F$ with respect to ϵ and ϵ' , since only the states near the Fermi energy ϵ_F are involved in electron–phonon scattering. Thus, the electron–phonon couple parameter can be computed using^{15,29}

$$G = \frac{\pi \hbar k_B \lambda \langle \omega^2 \rangle}{g(\epsilon_F)} \int_{-\infty}^{\infty} g^2(\epsilon) \left(-\frac{\partial f}{\partial \epsilon} \right) d\epsilon \approx \pi \hbar k_B g(\epsilon_F) \lambda \langle \omega^2 \rangle \quad (7)$$

where λ is the electron–phonon mass enhancement parameter,³⁰ and $\langle \omega^2 \rangle$ is the McMillan Factor for the second moment of the phonon spectrum.³¹ The last approximation is based on the fact that $-\partial f / \partial \epsilon \approx \delta(\epsilon - \epsilon_F)$ at room temperature. Using the approximate relation $\langle \omega^2 \rangle \approx \theta_D^2 / 2$,^{31,32} the mass enhancement parameter $\lambda_{Mo} = 0.42$,^{33,34} and the electron DOS at Fermi level of 0.61 states/(eV·cell),³⁴ the above equation yields $G = 1.2 \times 10^{17}$ W/m³ K for Mo at room temperature.

The thermal conductance across the Mo–Si interface, $h_{B,Mo-Si}$ can be derived using the method proposed by Majumdar and Reddy⁴ as

$$h_{B,Mo-Si}^{-1} \approx \frac{1 + \sqrt{Gk_{p,Mo}} / h_{pp,Mo-Si}}{\sqrt{Gk_{p,Mo}}} = h_{ep}^{-1} + h_{pp,Mo-Si}^{-1} \quad (8)$$

where $k_{p,Mo}$ is the phonon thermal conductivity of Mo. This equation uses a serial connection of the thermal resistances from electron–phonon nonequilibrium ($R_{ep} = 1/h_{ep}$) and interface phonon scattering ($R_{pp} = 1/h_{pp,Mo-Si}$). The DMM

evaluates the phonon–phonon conductance h_{pp} and is discussed in more detail in the Supporting Information. Equation 8 assumes that the electron and phonons are in equilibrium far away from the interface. One problem with this assumption arises when the Mo film thickness becomes close to the electron cooling length, $L_c = [Gk_{Mo}/(k_{Mo,e}k_{Mo,p})]^{1/2} = 1.4$ nm,²⁷ resulting in strong electron–phonon nonequilibrium and incomplete convergence of T_e and T_p . We have performed numerical simulations using the phonon Boltzmann transport equation yielding thermal resistance comparable with that predicted by the TTM for the 2.8 nm film thickness, with a difference less than 30%. Therefore, the TTM provides reasonable estimates of the electron–phonon nonequilibrium for the thicknesses our Mo/Si multilayers. More details on the comparison are provided in the Supporting Information.

Phonons in polycrystalline metal films can conduct heat by traveling directly across the film, either ballistically or impaired by scattering predominantly with each other and with defects, without transferring significant energy to the electron system. Although this conduction path is usually negligible in thick metal films due to the small phonon thermal conductivity,¹⁵ the extra burden of electron–phonon energy exchange imposed on the electron system can render direct phonon conduction the dominant mechanism. In our Mo/Si multilayer samples, the direct phonon transport path experiences a thermal resistance $R_p = d_{Mo}/k_{p,Mo}$ which is in parallel with the electron path R_e as shown in Figure 3a. The phonon thermal conductivity of Mo can be estimated by

$$k_{p,Mo} = \frac{1}{3} (C_v v \lambda)_p \quad (9)$$

where C_v is the volumetric phonon heat capacity estimated using the Debye approximation,²⁶ v is the speed of sound, and λ is the phonon mean free path in Mo which takes into account the thin film size effect. Although the spatial confinement from the thin film thickness can modify the phonon properties, such as group velocity, polarization, density of states, etc., this expression provides an estimate of k_p when taking the size effect into the calculation of λ through Matthiessen's rule.²⁶ Figure 3b shows that R_p monotonically decreases with decreasing Mo thickness and crosses R_e at a “dominant carrier transition thickness”, which can be derived as $d_{trans} = 2k_e k_p / [(Gk_p)^{1/2} (k_e - k_p)]$. The phonon path starts to dominate the heat conduction

in ultrathin films below d_{trans} . The overall thermal resistance of a single Mo/Si repetition now becomes

$$R_{\text{Mo/Si}} = \frac{d_{\text{Mo}} + d_{\text{Si}}}{k_{\text{tot},z}} = R_{\text{Mo}} + R_{\text{Si}} + R_{\text{b,Mo-Si}} + R_{\text{b,Si-Mo}}$$

$$= \frac{d_{\text{Si}}}{k_{\text{Si},z}} + (R_{\text{ei}} \parallel R_{\text{p}}) + \frac{1}{h_{\text{pp,Mo-Si}}} + \frac{1}{h_{\text{pp,Si-Mo}}} \quad (10)$$

The TTM discussed earlier assumes a diffusive process in which the electrons reflect elastically off the interface and eventually thermalize the phonons in the metal side. However, the electrons can also scatter inelastically at the interface and exchange energy with the phonon system in the dielectric. This creates an additional path for thermal transport which is more important when the metal film thickness is close to or less than the electron mean free path, although it was largely ignored in the previous modeling of ultrathin metal/dielectric multilayers.^{1,2,35} Past experiments have shown evidence of the inelastic heat loss from a metal film to a dielectric using the transient thermoreflectance (TTR) technique.^{13,24,36} Several theoretical models also made an effort to quantify the inelastic thermal conductance including the inelastic phonon radiation limit theory,³⁷ the maximum transmission model,³⁸ and the anharmonic inelastic model.³⁹ These methods either provide a strict upper limit for the interface conductance or require the detailed dispersion relationship. In this paper we employ the model developed by Sergeev^{40,41} which uses a quasi-analytical approach combined with experimental data. The thermal boundary conductance due to inelastic electron-interface scattering can be written as

$$h_{\text{ei}} = \frac{\pi^4}{10} \frac{g(\epsilon_{\text{F}}) T^3}{p_{\text{F}}^2} \left(\frac{\beta_{\text{l}}}{v_{\text{l}}} + 2 \frac{\beta_{\text{t}}}{v_{\text{t}}} \right) J \left(\frac{T}{\theta_{\text{D}}} \right) \quad (11)$$

where θ_{D} is the Debye temperature, p_{F} is the Fermi momentum, $g(\epsilon_{\text{F}})$ is the electron DOS at the Fermi level, and v_{l} and v_{t} are the longitudinal and transverse sound velocities, respectively. The integral term $J(y) = -(15/4\pi^4) \int_0^y x^4 \nabla N(x) dx$ expands the formula into the high-temperature regime. The interaction constants of electrons with longitudinal and transverse phonons, β_{l} and β_{t} , are determined by fitting the experimental data of Nb films, as in Table 2.⁴⁰

Table 2. Material Properties of Molybdenum Used in the Calculation^a

v_{l} [10^3 m/s]	v_{t} [10^3 m/s]	v_{F} [10^6 m/s]	β_{l}	β_{t}	τ_{e} [10^{-15} s]
6.25	3.35	1.7	10	35	3.80

^aThe interaction constants β_{l} and β_{t} are obtained by fitting the data of Nb films.⁴⁰ The Fermi velocity v_{F} and electron relaxation time τ_{e} of Mo are taken from ref 43.

Since Nb and Mo have nearly the same electron orbital behavior and electronic band structures,⁴² we assume the electron scattering parameters of Mo are similar to those of Nb. Future measurements on the temperature-dependent electrical resistivity of Mo film will provide more accurate β_{l} and β_{t} . The inelastic electron scattering at interface adds another conduction path in parallel to the electron and phonon paths in a more complex fashion, as in Figure 4. A portion of the electrons in the Mo layer transfer their energy through electron–phonon coupling resistance R_{ep} (by TTM) and subsequently the

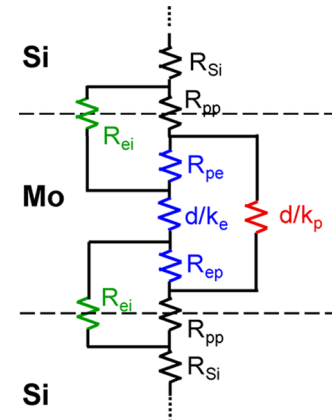


Figure 4. Complete thermal resistor network of the metal/semiconductor multilayers. Three physical mechanisms are included: electron–phonon nonequilibrium (R_{pe} and R_{ep}), direct phonon transport (d/k_{p}), and inelastic electron-interface scattering (R_{ei}).

phonon boundary resistance R_{pp} , while the other electrons can scatter inelastically at the Mo–Si interface and lose energy to the Si lattice through R_{ei} . The effective cross-plane thermal conductivities of the Mo/Si multilayers computed from this model agree reasonably well with the experimental data for various thickness ratios ($d_{\text{Mo}}/d_{\text{total}}$) as shown in Table 1.

In conclusion, for heat conduction normal to dielectric–metal interfaces, nonequilibrium between electrons and phonons prevails in the metal near the interface within a distance comparable to the phonon mean free path. In metal/dielectric multilayers in which the metal film thickness is only a few nanometers, the direct phonon conduction across the thin metal layer can present a dramatically more effective heat-transfer path than the electron conduction path that dominates in thicker metals. By comparing the electron and phonon resistances developed in the network model in Figure 3a, an approximate transition thickness $d_{\text{trans}} = 2k_{\text{p}}k_{\text{e}}/[(Gk_{\text{e}}k_{\text{p}})^{1/2}(k_{\text{e}} - k_{\text{p}})]$ can be extracted for the transition from electron to phonon dominated conduction. This criterion neglects the interface scattering and temperature dependence of the conductivities but captures the main physical arguments developed here. As the metal film thickness shrinks below the electron mean free path, more electrons can travel ballistically and scatter inelastically at the metal–dielectric interface, adding another energy-transfer path from the electron system in the metal to the phonon system in the dielectric. The thermal network model (Figure 4) considers all three conduction paths: the electron–phonon nonequilibrium (R_{ep} and R_{pe}), direct phonon transport (R_{p}), and inelastic electron-interface scattering (R_{ei}). The theoretical calculations agree with the measured thermal conductivities of two Mo/Si multilayers samples with period of 6.9 nm and Mo layer thickness fractions of 0.4 and 0.6. A more rigorous study based on the electron–phonon coupled, two-dimensional Boltzmann transport equation may provide a more detailed view of the multiple conduction mechanisms and their relative strengths.

■ ASSOCIATED CONTENT

Supporting Information

Additional TEM images of the Mo/Si multilayers, detailed discussion on the diffuse mismatch model for phonon–phonon interface conduction, and a numerical verification of the thermal resistor network using the Boltzmann transport

equation. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest.

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