Thermal Boundary Resistance Effect on Non-Equilibrium Energy Transport in Metal-Dielectric Thin Films Heated by Femtosecond Pulse Lasers

Jae Bin Lee and Seong Hyuk Lee*

School of Mechanical Engineering, Chung-Ang University, 221 Heuksuk-dong, Dongjak-gu, Seoul 156-756, Korea

The aim of this study is to investigate the effect of interfacial thermal boundary resistance (TBR) at a metal-dielectric interface on non-equilibrium energy transport in Au/SiO₂ films heated by femtosecond pulse lasers. In this paper we suggest a combined set of numerical models that include the two-temperature model (TTM) for a metal side and the heat conduction equation for a dielectric layer. In addition, the TBRs between metal and nonmetal layers are calculated using thermal conductance, which is closely associated with the electron-phonon resistance and phonon-phonon resistance. Herein we present the transient and spatial distributions of TBR for a Au/SiO₂ film irradiated by a 100-fs pulse laser with a 1053 nm wavelength, which are substantially affected by the phonon temperature and electron-phonon coupling. We also discuss the effect of laser fluence on the TBR and energy transport. The TBR rapidly increases the thermal conductivity at the interface, and becomes dominant at an early stage of laser irradiation over a very short period and then drastically decreases with time. Moreover, the TBR is substantially affected by the electron-phonon coupling and it should be considered for more accurate prediction of the lattice temperature drop at the interface. [doi:10.2320/matertrans.M2011021]

(Received January 13, 2011; Accepted April 7, 2011; Published June 25, 2011)

Keywords: femtosecond pulse laser, metal-dielectric interface, thermal boundary resistance (TBR), electron-phonon coupling factor, non-equilibrium, thin film

1. Introduction

In recent years, femtosecond pulse lasers have been regarded as a promising technique in material processing because the thermal affected zone and collateral damage can be substantially reduced due to the time scale difference between the pulse duration and heat diffusion time. Much effort has been made in a wide variety of fields including medical applications,¹⁾ material processing,²⁾ molecular spectroscopy,³⁾ ionization and dissociation of polyatomic molecules.⁴⁾ For such applications of ultra-short pulse lasers, it is necessary to investigate the non-equilibrium process between energy carriers, fast phase transition, theoretical modeling, and the non-linear effect on laser ablation.

Currently, considerable effort has been devoted to the theoretical and experimental study of non-equilibrium energy transport in metallic thin films. Among many theoretical models for femtosecond pulse laser interactions with metals, the two-temperature model (TTM) has been widely used to examine electron-phonon interactions and non-equilibrium energy transport in metallic thin films.^{5–12)} Because the electron-electron interaction time (on the order of a hundred femtoseconds) is shorter than the electron-phonon interaction time (on the order of picoseconds), a strong non-equilibrium occurs between the electrons and phonons under the irradiation of femtosecond laser pulses. Thermal equilibrium among the excited electrons is reached within a few hundred femtoseconds, depending on the excitation energy above the Fermi level.¹³⁾ Cooling of the electrons occurs because of the electron-phonon energy transport and electronic diffusive motion.

Multilayer thin films have been used to make useful and applicable structures such as superconductors, microelectronic layer packages,¹⁴⁾ and multi-layer metal films.^{15,16)}

Generally, the existence of thermal boundary resistance (TBR) at the interface between dissimilar films would significantly affect the device performance, and the importance of TBR has been recognized with the development of micro-electromechanical systems (MEMS).¹⁴⁾ For these reasons, research reporting the detailed mechanism of the TBR phenomena at solid-solid interfaces has been conducted.^{17–21)} Most researchers have used the diffuse mismatch model (DMM) to describe solid-solid interfacial TBR. This model considers phonon transmission across the interface under the assumption that the electron-phonon coupling resistance can be neglected.¹⁸⁾ Majumdar and Reddy²⁰⁾ suggested the TBR model, which accounts for the phonon-phonon scattering resistance at the interface between metal and nonmetal and the electron-phonon coupling resistance in metals. TBR causes a rapid decrease in the lattice temperature, resulting in thermal damage of solids.

Although abundant research has been performed, the interaction of a femtosecond pulse laser with multilayer metal-dielectric films is poorly understood, and TBR modeling has not been applied to ultra-short pulse laser interaction with multilayer film structures. Moreover, it is impossible to measure the *in situ* profile of the interfacial resistance due to the very short time scale and because the interfacial layer cannot be determined experimentally. Hence, the effect of TBR on energy transport in metal-dielectric multilayer films irradiated by a femtosecond pulse laser requires further investigation. Thus, the main objective of this study was to numerically investigate the effect of the interfacial TBR in metal-dielectric (Au/SiO₂) thin films heated by femtosecond pulse lasers on non-equilibrium energy transport. Prior to the primary simulation, we validated our computational code by comparing the electron temperatures predicted for an Au single film with previous numerical results and experimental data. We suggest a combined set of energy equations, which include two-temperature equations for a metal side and a

*Corresponding author, E-mail: shlee89@cau.ac.kr

well-known conduction equation for a dielectric side. The transient evolution of TBR estimated at the interface is also examined in this article, particularly the analysis of the close interaction of TBR with the phonon temperature and electron-phonon coupling.

2. Numerical Modeling and Computational Details

2.1 A combined computational model of metal-dielectric thin films

According to Majumdar and Reddy,²⁰⁾ the energy transport across metal-dielectric interfaces must occur between electrons and phonons. There are two different energy transport pathways: (1) coupling between electrons of the metal and phonons of the dielectric at the interface; and (2) coupling between electrons and phonons within the metal. We suggest a combination method to numerically simulate a metal-dielectric thin film irradiated by ultra-short pulse lasers. The two-temperature model (TTM) has been widely used to simulate the electron-phonon interaction for metals irradiated by an ultra-short pulse laser. Because a focused laser beam diameter is typically much larger than the penetration depth of the metal, one-dimensional analysis is valid, and the spatial and temporal evolutions of the electron and lattice temperatures are expressed as follows:^{7,12)}

$$\frac{\partial C_e(T_e)T_e}{\partial t} = \frac{\partial}{\partial z} \left(k_e(T_e) \frac{\partial T_e}{\partial z} \right) - G(T_e - T_l) + S(z, t) \quad (1)$$

$$\frac{\partial C_l(T_l)T_l}{\partial t} = G(T_e - T_l) \quad (2)$$

$$S(z, t) = \frac{0.939F(1 - R(t))}{t_p} \exp\left(-\int_0^z \alpha(z, t)dz\right) \times \exp\left(-2.773 \frac{t^2}{t_p^2}\right), \quad (3)$$

where $k_e(T_e)$ is the electron thermal conductivity, G is the electron-phonon coupling factor, and $S(z, t)$ is the laser heating source term. In eq. (3), $R(t)$, F , t_p , and $\alpha(z, t)$ represent the reflectivity at the surface, laser fluence, pulse duration, and absorption coefficient, respectively. In TTM, some of the thermal and optical parameters should be carefully modeled to ensure more accurate predictions. First, the electron heat capacity is commonly expressed as a linear function of the electron temperature, $C_e(T_e) = \gamma T_e$, where $\gamma = \pi^2 n_e k_B^2 / 2\varepsilon_F$.²²⁾ Because this Sommerfeld expansion is no longer valid for high electron temperatures, the electron heat capacity is calculated from the following equation:¹³⁾

$$C_e(T_e) = \int_{-\infty}^{\infty} \frac{\partial f(\varepsilon, \mu, T_e)}{\partial T_e} g(\varepsilon) \varepsilon d\varepsilon, \quad (4)$$

where $g(\varepsilon)$ is the electron density of states (DOS), μ is the chemical potential at T_e , and $f(\varepsilon, \mu, T_e)$ is the Fermi distribution function. Based on the Drude theory,²³⁾ we calculated the electron thermal conductivity, which can be expressed as

$$k_e(T_e) = \frac{1}{3} C_e(T_e) V_e^2(T_e) \tau_e^*(T_e), \quad (5)$$

where V_e denotes the electron velocity, and τ_e is the electron-electron relaxation time with respect to the electron temper-

ature.²³⁾ It is noted that for τ_e related to the collision frequency, there are three different regimes depending on the melting temperature and Fermi temperature: the cold solid regime (i), hot plasma regime (ii), and intermediate regime (iii), which correspond to the following expressions:^{9,24)}

$$(i) \quad v = k_s \left(\frac{m_e}{M} \right)^{1/2} \frac{J_i}{\hbar} \frac{T_l}{T_D} (T_e < T_{\text{melt}}) \quad (6)$$

$$(ii) \quad v = k_s \frac{Z n_e \ln \Lambda}{T_e^{3/2}} (T_e \geq T_F) \quad (7)$$

$$(iii) \quad v = k_s \frac{V_e}{r_0} (T_{\text{melt}} < T_e < T_F), \quad (8)$$

where the empirical constant k_s can be obtained from experimental data using the interfacial condition.²⁴⁾ Generally in thin film optics, the reflectivity and absorption coefficient can be expressed as $R = ((n - 1)^2 + k^2) / ((n + 1)^2 + k^2)$ and $\alpha = 4\pi k / \lambda$, respectively, and they are not constant values during laser irradiation. The complex refractive index can be expressed by $n = ((\varepsilon_1 + (\varepsilon_1^2 + \varepsilon_2^2)^{1/2}) / 2)^{1/2}$ and $k = ((-\varepsilon_1 + (\varepsilon_1^2 + \varepsilon_2^2)^{1/2}) / 2)^{1/2}$ where n and k represent the refractive index and the extinction coefficient, respectively.⁹⁾ The Drude model was used to determine the complex dielectric function, $\varepsilon_1 + i\varepsilon_2$, which is described by $\varepsilon_1 = 1 - (\omega_p^2 / (\omega_p^2 + \nu^2))$ and $\varepsilon_2 = \nu \omega_p^2 / (\omega_L^3 + \nu^2 \omega_L)$.^{9,24)} Thus, optical characteristics depend highly on the collision frequency, suggesting variation in the reflectivity during laser irradiation.

The electron-phonon coupling factor, G , should be carefully chosen in metals because electron-phonon coupling plays a major role in transferring electron energy to the lattice phonon. Previously, Lee *et al.*²⁵⁾ compared theoretical models for electron-phonon coupling in a single Au film irradiated by a femtosecond pulse laser. They showed that electron-phonon coupling behavior depends significantly on the laser fluence and should be carefully determined when describing non-equilibrium energy transport. For the present study, we used the theoretical model proposed by Lin *et al.*¹³⁾ to ensure consideration of electron scattering away from the Fermi surface at high electron temperatures. The rigorous form of temperature-dependent electron-phonon coupling can be expressed as:¹³⁾

$$G(T_e) = \frac{\pi \hbar k_B \lambda \langle \omega^2 \rangle}{g(\varepsilon_F)} \int_{-\infty}^{\infty} g^2(\varepsilon) \left(-\frac{\partial f}{\partial \varepsilon} \right) d\varepsilon, \quad (9)$$

where $g(\varepsilon)$ is the electron DOS at the energy level ε . The above equation reflects a major variation in the G -factor at high electron temperatures, whereas it requires some difficult procedures for electronic structure calculation of the electron DOS.

Similar to the previous work of Ashcroft and Mermin,²⁶⁾ we used the conventional heat conduction model in a SiO₂ layer to calculate the lattice phonon temperature on a dielectric side in the present study. This approach is valid under our conditions of laser irradiation in which electrons in the valance band of SiO₂ are not excited beyond a band gap energy of 9.0 eV for SiO₂.^{18,26–28)} Therefore, the energy transport via electrons can be neglected in a SiO₂ layer where conductive heat transfer becomes dominant. We used the simple conduction equation for a dielectric SiO₂ layer:

$$\rho_{\text{sm}} \frac{\partial C_{\text{sm}} T}{\partial t} = \frac{\partial}{\partial z} \left(k_{\text{sm}} \frac{\partial T}{\partial z} \right), \quad (10)$$

where ρ_{sm} , C_{sm} , and k_{sm} are the density, heat capacity, and thermal conductivity of SiO_2 , respectively.

2.2 Thermal boundary resistance (TBR) and numerical details

Many researchers have used the acoustic mismatch model (AMM) or the diffuse mismatch model (DMM) for TBR at a solid-solid interface.^{29–31} In these previous studies, AMM and DMM considering only the phonon-phonon resistance were used for energy transfer at the metal-nonmetal interface under the assumption that electron-phonon coupling resistance can be ignored. However, Majumdar and Reddy²⁰ suggested electron-phonon coupling resistance at the interface by assuming that in the metal, $k_e \gg k_p$ such that the harmonic mean of the electronic and lattice thermal conductivities is equal to k_p . The simplified relation of the interfacial conductance can be written as

$$h \approx \frac{\sqrt{Gk_p}}{\left[1 + \frac{\sqrt{Gk_p}}{h_{\text{pp}}} \right]} = \frac{h_{\text{ep}} h_{\text{pp}}}{h_{\text{ep}} + h_{\text{pp}}}, \quad (11)$$

where h_{ep} can be expressed as $(Gk_p)^{1/2}$, and is the electron-phonon thermal conductance in metal.²⁰ Additionally, k_p denotes the thermal conductivity in metal and h_{pp} indicates the phonon-phonon thermal conductance per unit area that can be predicted using DMM. According to Majumdar and Reddy,²⁰ at sufficiently low temperatures, h_{pp} can be expressed by ξT^3 , where T denotes the lattice temperature on the dielectric side and ξ is the empirical constant. In the present calculation, the empirical constant ξ is assumed to be unity at phonon temperatures less than the melting temperature of SiO_2 . At a higher phonon temperature, the role of electron-phonon coupling becomes more important, and even dominant in certain high temperature regimes.²⁰ As shown in eq. (11), the TBR is closely associated with the electron-phonon coupling factor, which is substantially dependent on the electron energy excited by the femtosecond pulse laser. Equation (11) includes two resistances: electron-phonon coupling resistance ($R_{\text{ep}} = 1/h_{\text{ep}}$) and phonon-phonon resistance at the interface ($R_{\text{pp}} = 1/h_{\text{pp}}$). The interfacial thermal conductivity k_{in} can be determined by hL , where L is the characteristic length.

For discretization of the governing equations (see eqs. (1) and (2)) in the present study, the finite difference method (FDM) was used with the fully implicit scheme. For the diffusion terms, a central differencing scheme with second order accuracy was used, and integration of the transient terms was performed over the control volume and time interval from t to $t + \Delta t$. The algebraic discretized equations were solved by the strongly implicit procedure (SIP).^{32,33} Figure 1 shows schematics of the Au/ SiO_2 multilayer structure irradiated by femtosecond pulse lasers. d_f , d_s , and d_{in} denote the depths of the Au film, SiO_2 substrate, and interface, respectively. Additionally, $T_{\text{int,top}}$ and $T_{\text{int,bottom}}$ are the temperatures at the top and bottom surfaces of the interface region, respectively.

Fig. 1 Schematics of the Au/ SiO_2 film structure and the corresponding interface region between the two dissimilar films.

The von Neumann boundary conditions with zero gradient at $z = 0$ and $z = L$ were used for the temperatures of the electrons and lattice phonons under the assumption that during a short period of laser heating, heat loss at the top and bottom surfaces is neglected.³⁴ The boundary conditions are as follows:

$$\left. \frac{\partial T_e}{\partial z} \right|_{z=0} = \left. \frac{\partial T_l}{\partial z} \right|_{z=0} = 0, \quad (12a)$$

$$\left. \frac{\partial T_e}{\partial z} \right|_{z=L} = \left. \frac{\partial T_l}{\partial z} \right|_{z=L} = 0. \quad (12b)$$

In addition, the electron and phonon temperatures were initially set at 300 K, indicating that the electrons and phonons are initially in a thermal equilibrium state. As shown in Fig. 1, there is an interface layer with a finite depth between two dissimilar films. The depth of the interface layer is very difficult to measure accurately because of non-uniformity and a very small scale (on the order of a few nanometers). An interface thickness of one nanometer was thus assumed in the present study, and only one computational node was located at the center of the interface region to numerically couple the metal and dielectric sides. A nanometer interface thickness was determined from the preliminary tests. As shown in Fig. 1, the interfacial boundary condition of the electron temperature at $z = z_{\text{int}1}$, can be written as

$$k_e \left. \frac{dT_e}{dz} \right|_{z=z_{\text{int}1}} = 0. \quad (13)$$

The insulation condition for electrons in the metal side was used in the present study because of the absence of electrons in the dielectric layer. This condition is valid when the transferred electron energy does not exceed 9.0 eV, which is the band gap energy of the SiO_2 layer, indicating that the phonon energy transport becomes dominant at the interface between the metal and dielectric layers. Meanwhile, for the lattice phonons, the continuity of the phonon energy flux at both sides of the interface gives

$$k_p \left. \frac{dT_p}{dz} \right|_{z=z_{\text{int}1}^-} = k_{\text{in}} \left. \frac{dT_p}{dz} \right|_{z=z_{\text{int}1}^+}, \quad (14)$$

$$k_{\text{in}} \left. \frac{dT_p}{dz} \right|_{z=z_{\text{int}2}^-} = k_p \left. \frac{dT_p}{dz} \right|_{z=z_{\text{int}2}^+}, \quad (15)$$

Table 1 Thermal and optical properties of the Au film and SiO₂.

	Description	Value	Reference
Au	Electron number density, n_e	$5.9 \times 10^{28} [\text{m}^{-3}]$	Ref. 9, 26)
	Mass of ion, M	196.97 [a.m.u]	Ref. 23)
	Debye temperature, T_D	170 [K]	
	Melting temperature, T_{melt}	1337 [K]	Ref. 26)
	Fermi temperature, T_F	6.39×10^4 [K]	
	First ionization potential, J_i	9.22 [eV]	
SiO ₂	Density, ρ_{sm}	2.2 [g/cm ³]	
	Heat capacity constant, C_{sm}	740 [J/kg·K]	Ref. 27, 28)
	Thermal conductivity, k_{sm}	1.38 [W/m·K]	

where the positive and negative signs in the superscripts denote the downward and upward directions at the interface, as illustrated in Fig. 1. With an initial constant electron number density of $5.9 \times 10^{28} \text{ m}^{-3}$ ^{9,26)} in the Au film, the numerical simulations were conducted using eqs. (1), (2), and (10) and the initial and boundary conditions mentioned above. The converged solutions could be obtained when the residuals of the discretized equations were less than 10^{-6} . The physical properties of the Au film used in the present study and SiO₂ are listed in Table 1. In this study, numerical simulations were first conducted in order to evaluate the computational code for a 200-nm single Au film irradiated by a 100-fs pulse laser with a 1053-nm wavelength. After the code validation, we conducted extensive simulations for a metal-dielectric film structure that consisted of a bulk SiO₂ substrate with a 0.6 μm depth and a 100-nm Au film deposited onto the substrate. For all calculations, a 100-fs pulse laser with a 1053-nm wavelength was used, whereas a very low laser fluence of 0.001 J/cm^2 for a single film calculation was used for comparison with the experimental data. Laser fluence was varied from 0.01 to 0.3 J/cm^2 for calculations for the Au-SiO₂ film structure.

3. Results and Discussion

Prior to the main discussion, we attempt to examine the thermal responses of the physical properties depending on energy carriers because thermal properties and energy carriers are highly associated with the TBR during laser irradiation. Figure 2 shows the spatial distribution of the electron thermal conductivity that is closely related to the electron temperature. These results were obtained from the preliminary simulation for a 300-nm single Au film irradiated by a 100-fs pulse laser at a fluence of 0.1 J/cm^2 with a 1053-nm wavelength. The electron thermal conductivity increased at the top surface with the increase in electron heat capacity.^{9,24)} The electron thermal conductivity did not change at the bottom surface of the Au film due to the fact that the electron temperature was lower than the melting temperature. If the electron temperature is higher than 1337 K, the electron collision frequency lies in the hot plasma regime where electron conductivity gradually increases. The variation in electron thermal conductivity can directly affect energy transfer in gold film structures.²⁴⁾ For

Fig. 2 The spatial variation of the electron thermal conductivity of a 300-nm single Au layer exposed to a 100-fs pulse laser with 0.1 J/cm^2 irradiation.

validating our computational code, we conducted numerical simulations for a 200-nm Au film irradiated by a femtosecond pulse laser with $F = 0.001 \text{ J/cm}^2$ and $t_p = 100 \text{ fs}$. The predicted electron temperatures were compared with the numerical results reported previously by Sim and Lee²⁴⁾ and experimental results.³⁵⁾ As shown by the transient evolution of electron temperatures in Fig. 3, the change in normalized electron temperature was defined as $(T_e - T_{\text{eq}})/(T_e - T_{\text{eq}})_{\text{max}}$, which is analogous to the normalized reflectivity change and similar to literature.¹²⁾ As a result, our numerical predictions show good agreement with the experimental data and previous results.²⁴⁾

Even when previous researchers assumed that the interface layer was very thin,¹⁸⁾ they were unable to suggest a suitable thickness and their estimations were on the order of a few nanometers.^{18,29,36)} In the present study we created an interfacial region between the Au film and SiO₂ substrate for convenient modeling of a metal-nonmetal interface region, as shown in Fig. 1. Unfortunately, it is impossible to accurately determine a finite thickness of this interface region. Figure 4 shows the preliminary results of numerical

Fig. 3 Comparison of the electron temperatures among the numerical results from Sim and Lee,²⁴⁾ experimental data from Bronson *et al.*,³⁵⁾ and the present predictions at the top surface of a 200-nm single Au film after 0.001 J/cm² irradiation.

Fig. 4 Comparison of the variation of lattice temperature according to different interface depths (0.5, 1.0, 1.5, and 2.0 nm) at 1 ps and 5 ps after irradiation on Au/SiO₂ multilayer film (at $F = 0.01$ J/cm² and $t_p = 100$ fs).

sensitivity for various thicknesses of the interface region. The lattice phonon temperature distributions are shown at 1.0 and 5.0 ps after fs-laser irradiation was completed. At $t = 1.0$ ps, a substantial temperature difference was observed at the interface, indicating the existence of TBR. Alternatively, the TBR effect was drastically reduced at $t = 5.0$ ps. In particular, the differences among the lattice temperatures estimated for different thicknesses of the interface region were very small, and the maximum difference was less than approximately 5.0 K, suggesting that the numerical dependency on the thickness of the interface region can be sufficiently neglected. Therefore, the thickness of the interface region was set at one nanometer for all calculations.

Figure 5 predicts the change in electron temperature in the depth direction at different times. The electron temper-

Fig. 5 The change of electron temperature in the depth direction at different times (1, 2, 5, and 10 ps).

ature at the top surface rapidly increased during laser irradiation. After the increase, the temperature slowly decreased because of electron-phonon energy transport and electron-electron thermal conduction. The electron temperature at the interface gradually increased due to electron-electron thermal conduction. For electrons in dielectric materials (electrical insulators) to be excited, they must have absorbed energy greater than the 9.0 eV band gap energy of SiO₂,^{27,28)} above which the electrons in the valence bands can move toward the conduction band. However, under the present conditions of laser irradiation, the electrons in the valence band of SiO₂ were not excited beyond 9.0 eV. For instance, at $F = 0.3$ J/cm², the maximum electron temperature at the interface was estimated to be 1.16 eV, which was much less than 9.0 eV.

Figure 6 compares the predicted spatial distributions of lattice temperatures for low and high laser fluences. The lattice temperature rapidly decreased at the interface due to the TBR, which is affected by the lattice temperature and electron-phonon coupling. Indeed, the TBR affects the rapid decrease in interfacial thermal conductivity. As shown in Fig. 6(a), a temperature difference between the top and bottom surfaces of the interface region for low laser fluence ($F = 0.01$ J/cm²) was maintained up to approximately 58% of the maximum lattice temperature estimated at $t = 5.0$ ps. Alternatively, at higher laser fluence ($F = 0.3$ J/cm²), the TBR effect can be neglected after $t = 5$ ps, as shown in Fig. 6(b). Based on these results, we can conclude that the TBR at the interface was dominant at the early stage of laser irradiation for a very short time, and then drastically decreased with time. Hence, the interfacial thermal conductivity increased with time and its magnitude was nearly the same as the thermal conductivity of SiO₂.

Majumdar and Reddy²⁰⁾ calculated the thermal conductance using a constant value of the electron-phonon coupling factor (so-called, G -factor), and reported that the existing TBRs were affected only by lattice temperature. Unlike the previous work,²⁰⁾ we attempted to estimate TBRs in the present study by considering a non-linear change in the

Fig. 6 Comparison of the change of the lattice temperature with respect to depth when the Au/SiO₂ multilayer irradiated by a femtosecond laser with low (0.01 J/cm²) and high laser fluences (0.3 J/cm²) with and without TBR effects.

G -factor during laser irradiation. This is a clear difference between our study and the previous work of Majumdar and Reddy,²⁰⁾ and it is very important as the interfacial thermal conductance is closely associated with the electron-phonon coupling. In addition, our results may be helpful in understanding the major role of TBR at the interfaces during a very short time period in which very high photon energy absorption occurs. Figure 7 illustrates the transient evolution of TBR, the lattice temperature of the interface region, G -factor, and electron temperature. For this case, a 100-nm Au film and a relatively higher laser fluence of 0.3 J/cm² were used for the simulation. In Fig. 7(a) it is apparent that as the laser fluence increased, the rate at which TBR was decreasing rapidly increased due to increased electron-phonon coupling. Hence, at higher laser fluence, the lattice temperature increased faster than when the laser fluence was lower. Eventually, the TBR reached a saturated value, which depended on the lattice temperature. It is evident in Fig. 7(b) that the G -factor rapidly increased with

Fig. 7 (a) The prediction of the variation of thermal resistance and lattice temperature with respect to time, and (b) transient behavior of electron temperature and electron-phonon coupling factor ($F = 0.3 \text{ J/cm}^2$, $t_p = 100 \text{ fs}$, $d_f = 100 \text{ nm}$).

the electron temperature of the Au film during laser irradiation. This result supports the fact that TBR is strongly dependent on the behavior of electron-phonon coupling and electron temperature.

Figure 8 shows the transient evolution of the temperature difference estimated at the top and bottom surfaces of the interface region. A large difference (approximately 150 K) exists between the results obtained with and without considering the TBR effect, indicating that the presence of TBR should be considered for more accurate prediction of the drop in lattice temperature occurring at the interface. At the early stage of laser irradiation, the temperature drop substantially increased at the interface region, and gradually decreased with time. Additionally, the maximum temperature difference is achieved more quickly with increasing laser fluence due to variation in the rates of lattice temperature increase in the Au film. Moreover, the maximum temperature difference of the interface region reached 32% of the maximum lattice temperature estimated at $t = 2.0 \text{ ps}$ for $F = 0.3 \text{ J/cm}^2$, indicating that the temperature drop at the interface region may cause thermal damage in a multilayered structure.¹⁴⁾

Fig. 8 The comparison of the magnitude of temperature difference with time at various laser fluences (0.01, 0.1, 0.3 J/cm²): (a) with and (b) without TBR effects.

4. Conclusions

In the present study we numerically investigated TBR effects on non-equilibrium energy transport in a multilayer Au/SiO₂ film structure irradiated by a femtosecond pulse laser. We established a combined set of energy equations to describe electron-phonon and phonon-phonon interactions and applied the thermal conductance model to Au/SiO₂ films heated by ultra-short pulse laser interactions. Based on our findings, the following conclusions can be drawn:

- (1) The TBR rapidly increased thermal conductivity at the interface. The temperature difference at the interface was maintained up to approximately 58% of the maximum lattice temperature at $t = 5.0$ ps for $F = 0.01$ J/cm², whereas the TBR did not affect the temperature difference after $t = 5.0$ ps for high laser fluence. The TBR at the interface was dominant at an early stage of laser irradiation for a very short time, and it drastically decreased with time.
- (2) The rate at which TBR was decreasing significantly increased due to the increased electron-phonon cou-

pling with the increase in laser fluence. At higher laser fluence, the lattice temperature increased faster than when the laser fluence was lower. Eventually, the TBR reached a saturated value, which depended on the lattice temperature.

- (3) A large difference in lattice temperature at the interface was observed and the lattice temperature difference reached 32% of the maximum lattice temperature estimated at $t = 2.0$ ps for $F = 0.3$ J/cm², indicating that the temperature drop in the interface region may cause thermal damage in a multilayered structure. The maximum temperature difference at the interface was reached more rapidly as the laser fluence increased due to variation in the rates of lattice temperature increase in the Au films.
- (4) In spite of a rapid increase in the lattice temperature of the Au film, the phonon energy was rarely transferred from the Au film to the SiO₂ substrate due to the existence of TBR. Additionally, TBR was strongly dependent on the behavior of electron-phonon coupling and electron temperature. Therefore, the presence of TBR should be considered to ensure more accurate prediction of the lattice temperature drop occurring at the interface.

Acknowledgements

This work was sponsored by the Human Resources Development of the Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea government, Ministry of Knowledge and Economy (2008-PEP-HM-E-05-0000).

REFERENCES

- 1) P. F. Sharp, A. Manivannan, H. Xu and J. V. Forrester: *Phys. Med. Biol.* **49** (2004) 1085–1086.
- 2) I. M. Burakov, N. M. Bulgakova, R. Stoian, A. Rosenfeld and I. V. Hertel: *Appl. Phys. A* **81** (2005) 1639–1645.
- 3) K. Claytor, S. Khatua, J. M. Guerrero, A. Tcherniak, J. M. Tour and S. Link: *J. Chem. Phys.* **130** (2009) 164710–164719.
- 4) X. Zhang, D. D. Zhang, H. Liu, H. F. Xu, M. X. Jin and D. J. Ding: *J. Phys. B* **43** (2010) 025102–025108.
- 5) H. S. Sim, K. G. Kang, S. H. Lee, J. M. Kim and Y. E. Shin: *Mater. Sci. Forum* **580–582** (2008) 143–146.
- 6) K. G. Kang, S. H. Lee, H. S. Ryou, Y. K. Choi, S. H. Park and J. S. Lee: *Mater. Trans.* **49** (2008) 1880–1888.
- 7) S. I. Anisimov, B. L. Kapeliovich and T. L. Terel'man: *Sov. Phys. JETP* **39** (1974) 375–377.
- 8) J. K. Chen and J. E. Beraun: *J. Opt. A: Pure and Appl. Opt.* **5** (2003) 168–173.
- 9) K. Eidmann, J. Meyer-ter-Vehn, T. Schlegel and S. Huller: *Phys. Rev. E* **62** (2000) 1202–1214.
- 10) K. Furusawa, K. Takahashi, H. Kumagai, K. Midorikawa and M. Obara: *Appl. Phys. A* **69** (1999) S359–366.
- 11) L. Jiang and H. L. Tsai: *Proc. ASME Heat Transfer Summer Conference*, (San Francisco, CA, USA, 2005) No. HT2005-72199.
- 12) T. Q. Qui and C. L. Tien: *ASME J. Heat Transfer* **115** (1993) 835–841.
- 13) Z. Lin, L. V. Zhigilei and V. Celli: *Phys. Rev. B* **77** (2008) 075133.
- 14) K. Ramadan and M. A. Al-Nimr: *Int. J. Thermal Sci.* **48** (2009) 1718–1727.
- 15) W. M. G. Ibrahim, H. E. Elsayed-Ali, C. E. Bonner and M. Shinn: *Int. J. Heat Mass Transfer* **47** (2004) 2261–2268.
- 16) A. M. Chen, H. F. Xu, Y. F. Jiang, L. Z. Sui, D. J. Ding, H. Liu and M. X. Jin: *Appl. Surf. Sci.* **257** (2010) 1678–1683.

- 17) G. Chen: J. Heat Trans. **119** (1997) 220–229.
- 18) H. C. Chien, D. J. Yao and C. T. Hsu: Appl. Phys. Lett. **93** (2008) 231910.
- 19) P. E. Hopkins, J. L. Kassebaum and P. M. Norris: J. Appl. Phys. **105** (2009) 023710.
- 20) A. Majumdar and P. Reddy: Appl. Phys. Lett. **84** (2004) 23.
- 21) E. S. Landry and A. J. McGaughey: Phys. Rev. B **80** (2009) 165304.
- 22) C. L. Tien, A. Majumdar and F. M. Gerner: *Microscale Energy Transport*, (Taylor & Francis 1998).
- 23) C. Kittel: *Introduction to Solid State Physics*, (John Wiley & Sons 1986).
- 24) H. S. Sim and S. H. Lee: Numerical Heat Transfer Part A **54** (2008) 279–292.
- 25) J. B. Lee, K. G. Kang and S. H. Lee: Mater. Trans. **52** (2011) 547–553.
- 26) N. W. Ashcroft and N. D. Mermin: *Solid State Physics*, (Thomson Learning, 1976).
- 27) M. J. Weber: *Handbook of optical materials*, (CRC Press Laser and Optical Science and Technology Series 2003).
- 28) G. Neudeck: The PN Junction Diode, (Addison-Wesley Publishing Co. 1989).
- 29) D. G. Cahill, W. K. Ford, K. E. Goodson, G. D. Mahan, A. Majumdar, H. J. Maris, R. Merlin and S. R. Phillpot: J. Appl. Phys. **93** (2003) 793–818.
- 30) Y. S. Ju, M. T. Hung and T. Usui: J. Heat Transfer **128** (2006) 919–925.
- 31) E. T. Swartz and R. O. Pohl: Rev. Mod. Phys. **61** (1989) 605–668.
- 32) S. V. Patankar: Numerical heat transfer and fluid flow, (Taylor & Francis, 1980).
- 33) H. K. Versteeg and W. Malalasekera: An introduction to computational fluid dynamics, (Longman, 1995).
- 34) S. H. Lee, H. S. Sim, J. Lee, J. M. Kim and Y. E. Shin: Mater. Trans. **47** (2006) 2835–2841.
- 35) S. D. Bronson, A. Kazeroonian, J. S. Moodera, D. W. Face, T. K. Cheng, E. P. Ippen, M. S. Dresselhaus and G. Dresselhaus: Phys. Rev. Lett. **64** (1990) 2173–2175.
- 36) W. L. Chan, R. S. Averback and D. G. Cahill: Appl. Phys. A **97** (2009) 287–294.

Nomenclature

C_e electron heat capacity, W/m³
 C_l lattice heat capacity, W/m³
 C_{sm} heat capacity of SiO₂, W/m³
 d_f film thickness, m
 d_{in} interface depth, m
 d_m multilayered structure thickness, nm
 d_s substrate thickness, m
 E_g band gap energy, J
 G electron-phonon coupling constant, W/m³K
 $g(\varepsilon)$ electron density of states (DOS), states/eV/atom
 $\hbar$ reduced Planck constant, Js
 h thermal conductance, W/m²K
 I laser intensity, W/m²
 F laser fluence, J/cm²
 J_i ionization potential, eV
 k extinction coefficient
 k_e electron thermal conductivity, W/mK
 k_l lattice thermal conductivity, W/mK
 k_B Boltzmann constant, Js
 k_s empirical constant
 k_{sm} thermal conductivity of SiO₂, W/mK
 L characteristic length, m

M ion mass, a.m.u
 m_e electron mass, kg
 n_i ion number density, /m³
 n_e electron number density, /m³
 n refractive index
 r_o ion sphere radius, m
 R reflectivity
 R_B thermal boundary resistance, K/W
 S laser heating source term, W/m³
 T lattice temperature on the nonmetal side, K
 T_e electron temperature, K
 T_l phonon (lattice) temperature on the metal side, K
 T_D Debye temperature, K
 T_F Fermi temperature, K
 $T_{int,top}$ temperature at the top surface of interface regime, K
 $T_{int,bottom}$ temperature at the bottom surface of interface regime, K
 T_{melt} melting temperature, K
 t time, s

t_p pulse duration time (FWHM), s

V_e electron velocity, m/s

z z -direction, m

Z ionization degree

Greek symbols

α linear absorption coefficient, /m
 β two-photon absorption coefficient, m/W
 ε average kinetic energy of electron, eV
 ε_1 real part of dielectric function
 ε_2 imaginary part of dielectric function
 $\hat{\varepsilon}$ dielectric function
 λ laser wavelength, m
 λ_e electron mean free path, m
 μ the chemical potential, J
 τ_e electron-electron relaxation time, s
 $\ln \Lambda$ Coulomb logarithm
 ν collision frequency, /s
 ρ_{sm} density of SiO₂, /m³
 ω_L laser frequency, /s
 ω_p plasma frequency, /s
 $\hbar$ reduced Planck constant, Js
 $\hbar\omega$ photon energy quantum

Superscripts

0 old values at time t

Subscripts

CV control volume
 e electron
 ep electron-phonon
 e* interface between nodes
 f films
 i ion
 l lattice
 int interface
 pp phonon-phonon
 s substrate