Effects of Temperature Coefficients for Dielectric Constants on Thermoreflectances and Thermal Responses of Metal Thin Films Exposed to Ultrashort Pulse Laser Beams

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Key words: Metal thin-film, Ultrashort pulse laser heating, Microscale heat transfer, Transient thermoreflectance, Complex dielectric constants

Abstract

Effects of temperature coefficients for dielectric constants on transient reflectances and thermal responses have been investigated for a metal (gold) thin-film during ultrashort pulse laser heating. Heating processes are simulated using the conventional conduction model (parabolic one-step, POS), the parabolic two-step model (PTS), the hyperbolic two-step model (HTS). Results from the HTS model are very similar to those from the PTS model, since the laser heating time in this study is considerably greater than the electron relaxation time. PTS and HTS models, however, result in completely different temperature profiles from those obtained by the POS model due to slow electron-lattice interactions compared to laser pulse duration. Transient reflectances are directly estimated from the linear relationship between electron temperature and complex dielectric constants, while conventional approaches assume that the change in reflectances is proportional to that in temperature. Reflectances at the front surface vary considerably for various dielectric constants, while those at the rear surface remain unchanged relatively.

Nomenclature

c : speed of light [m/s]
C : heat capacity [J/m^3 K]
G : electron-lattice coupling factor [W/m^3 K]
J : laser fluence [J/m^2]
k : thermal conductivity [W/m K]
k_o : wavenumber [1/m]
L : thin-film thickness [m]
n : real part of complex refractive index
q : heat flux [W/m^2]
R : reflectance
S : radiation absorption rate [W/m^3]
t : time [s]
t_c : electron thermalization time [s]
t_p : laser-pulse duration time [s]
T : temperature [K]

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v : temperature coefficient for dielectric constant
x : x-direction coordinate

Greek symbols

δ : radiation penetration depth [m]
ε : dielectric constant
χ : imaginary part of complex refractive index
τ_F : electron relaxation time [s]

Subscripts

0 : initial state
1, 2 : real and imaginary parts
e : electron
l : lattice

1. Introduction

Since its invention about 30 years ago, lasers have shown new directions in the fields of science and technology. Especially, they have contributed to develop many revolutionary methods to control spatial and temporal resolutions very precisely and thus make it possible to observe nonlinear optical phenomena, to control cell structures, to investigate very short time-scale phenomena, to synthesize noble materials, to manufacture microsystems, etc. The technology for these applications is based on the typical characteristics of lasers such as wavelength selectivity, high radiative intensity, and ultrashort pulse duration time.

In optics, the progress in temporal resolution was considerably slow compared to that in spatial resolution before the development of lasers. Since its development, however, the temporal resolution has been increased remarkably from microsecond up to femtosecond levels.

With the aid of ultrashort pulse lasers the very fast dynamics of free electrons in the metal films can be investigated, and thus, the electron-lattice thermalization mechanism could be understood and simplified to analyze the energy transfer process from photon absorption by electrons to internal energy dissipation through electron-lattice interactions.

Kaganov et al.[1] showed that lattice temperatures can be different from electron temperatures during the process shorter than electron relaxation times, which was confirmed experimentally.[2-3] Qiu and Tien[4] analyzed electron transports and electron-lattice interactions using the Boltzmann transport equation, simplified the scattering term in the equation through a rigorous quantum mechanical approach, and developed the simple theoretical model to describe the ultrashort time-scale phenomena, which is called the hyperbolic two-step (HTS) model.

The HTS model depicts the energy transfer process from photons of laser beam to lattice as follows: (1) photon absorption process by electrons, (2) energy transfer among electron gases, (3) energy transfer between electrons and lattices through the collision of electrons on lattices. In this model the characteristic time for each process is finite and thus, the electron-lattice interactions can be explored efficiently. This cannot be described by the one-step methods, whether they are parabolic (POS) or hyperbolic (HOS). They assume that electron-lattice interaction times are infinitesimally smaller than other processes.

Almost all the theoretical studies[2-4] assume that the reflection of the metal film during the ultrashort pulse laser heating remains unchanged. The actual reflectances,[2] however, can be changed by $\Delta R \sim 10^{-4}$ when the temperature changes by 10 K. When a metal thin-film is exposed to a very intense laser beam during very short time period, electron temperatures increase up to several thousand degrees and thus the change in surface reflectances becomes significant, which may influence the elec-

tron temperature rise in turn. In general, metal films of high reflectances are used for highly reflective surface materials of reflection mirrors. Although the absolute value of the change in reflectances may be small compared to the reflectances, it is not negligible in the respect of absorptances which control the film temperature rise.

The objective of this study, therefore, is to utilize the HTS model to analyze the thermal responses of metal films exposed to an ultrashort pulse laser beam and to understand the effect of transient thermoreflectance with the consideration of reflectance changes due to hot electrons.

2. Analysis

A schematic diagram is shown in Fig. 1 for the analysis on the thermal responses of metal thin-film exposed to an ultrashort pulse laser beam. The laser target in this study is a gold thin-film of thickness 100 nm, which is widely applied as coating materials of high reflection mirrors.

Since the response of metal films due to an ultrashort pulse laser beam is very short in time and very confined in space, it cannot be understood by the macroscopic diffusion models based on Fourier's law. This microscopic process can be described appropriately by the HTS model, which was derived by Qiu and

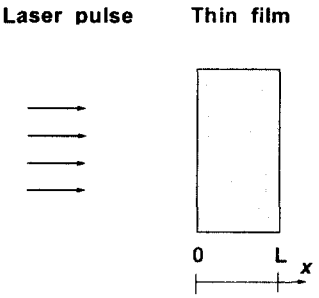


Fig. 1 Schematic diagram for ultrashort pulse laser heating of single metal layer films.

Tien[4] from the Boltzmann transport equation and will be applied in this study.

The thickness of the thin-film is usually very small (below 0.1 μm) and the penetration depth of the laser beam is relatively small (1 μm for 1 ns) in comparison to the diameter of the laser beam (above 10 μm). It, therefore, is assumed that the absorption process is transient and one-dimensional. The governing equations in this study are

$$C_e(T_e)\frac{\partial T_e}{\partial t} = -\frac{\partial q}{\partial x} - G(T_e - T_l) + S \quad (1)$$

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

$$\tau_F \frac{\partial q}{\partial t} + k(T_e, T_l)\frac{\partial T_e}{\partial x} + q = 0 \quad (3)$$

Here, the specific heat for electrons, C_e, is

$$C_e(T_e) = \frac{C_e^o T_e}{T_l} \quad (4)$$

since it is proportional to electron temperatures,[5] and the specific heat for lattice is assumed as a constant, since the lattice temperature in this study does not change much and is higher than the Debye temperature.[6] The thermal conductivity for electrons is proportional to free electron number density, electron relaxation time τ_F, and electron temperature. Since the relaxation time is assumed to be a constant due to the small changes in lattice temperature, the electron conductivity is simplified as,[7]

$$k(T_e, T_l) = \frac{T_e}{T_l} k \quad (5)$$

Equation (1) describes the photon absorption process by free electrons, heat diffusion among electron gas, and energy exchange between free electrons and phonons. Equation (2) is for

the energy exchange between phonons and electrons and the heat diffusion in the lattice is neglected since heat diffusion due to lattice vibration is much slower than that due to electron transport. Equation (3) is the microscopic wave heat transfer model for electron transport, rather than macroscopic model for phonon transfer.[8-10]

The radiation absorption term in Eq. (1) can be simplified as

$$S=0.94\frac{1-R}{t_p\delta}J\cdot\exp\left[-\frac{x}{\delta}-2.77\left(\frac{t-2t_p}{t_p}\right)^2\right] \quad (6)$$

where radiation absorption is assumed to decay exponentially and to be Gaussian temporally.[7] Here, R is the thin-film reflectance, t_p is the characteristic pulse duration time (full-width-half-maximum, FWHM), δ is the radiation penetration depth, and J is the laser fluence.

The objective of this study is to understand the effect of the transient thermoreflectance and thus, the reflectance should not be simplified as a constant. In general, radiation characteristics such as transmittances and reflectances are functions of the complex dielectric constant. The dielectric constant can be simplified as[11]

$$\varepsilon=\varepsilon_1(1+v_1T)+i\varepsilon_2(1+v_2T) \quad (7)$$

After the incidence of the laser beam on the film, the film temperature increases rapidly and thus, the temperature distribution inside the film becomes nonuniform. The change of the reflectance[11] in this case is

$$\frac{\Delta R}{R}=\frac{4\{[(n^2-x^2-1)I_2-2xnI_1]\varepsilon_1v_1+[(n^2-x^2-1)I_1+2xnI_2]\varepsilon_2v_2\}}{[(n+1)^2+x^2][(n-1)^2+x^2]} \quad (8)$$

where

$$\varepsilon_1=n^2-x^2 \quad (9)$$

$$\varepsilon_2=2xn \quad (10)$$

$$I_1=k_o\int_0^\infty T(x')\exp(-2k_oxx)\cos(2k_onx')dx' \quad (11)$$

$$I_2=k_o\int_0^\infty T(x')\exp(-2k_oxx')\sin(2k_onx')dx' \quad (12)$$

Here, $k_o=\omega/c$ is the wave number of the laser beam in vacuum. The initial and boundary conditions for electrons and phonons are

$$T_e(x,0)=T_l(x,0)=T_o \quad (13)$$

$$\left.\frac{\partial T_e}{\partial x}\right|_{x=0}=\left.\frac{\partial T_e}{\partial x}\right|_{x=L}=\left.\frac{\partial T_l}{\partial x}\right|_{x=0}=\left.\frac{\partial T_l}{\partial x}\right|_{x=L} \quad (14)$$

where heat losses from the front and back surface are assumed to be negligible.

Due to the nonlinearity the equations are solved numerically with the use of the Crank-Nicholson method, the central difference scheme and the iteration with convergence criteria of $\Delta T_e/T_e<10^{-4}$ and $\Delta T_l/T_l<10^{-5}$. During each time interval the difference between total energy absorbed in the film and the internal energy increase in the lattice and electrons is kept within 0.1%.

The properties and parameters used in this simulation are given in Table 1. Other than the

Table 1 Physical properties in this study[7,12]

Parameters	Estimated values
T_o	300 K
k	315 W/m K
C_l	2.5×10^6 J/m^3 K
C_{eo}	2.1×10^4 J/m^3 K
G	2.6×10^{16} W/m^3 K
L	100 nm
J	500 J/m^2
t_p	0.1 ps
δ	15.3 nm
τ_F	0.04 ps
λ	50.653 μm
$m=n+ix$	$0.166+3.15\,i$
v_1, v_2	$-1\times10^{-3}<v_1, v_2<1\times10^{-3}$ K^{-1}

temperature coefficients of the dielectric constant in Eq. (7), the properties in Table 1 are practical values.[7,12] Although there are no experimental data for the temperature coefficients, the coefficients are known to be in the range of $\pm 10^{-3}\,K^{-1}$.[13,14]

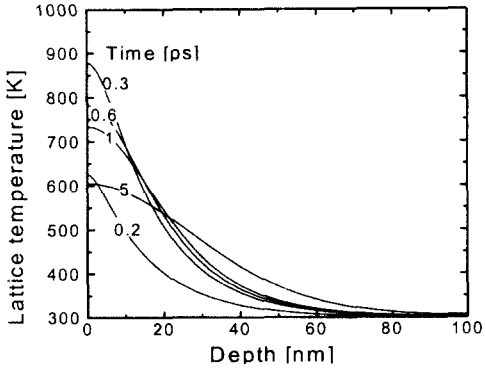
3. Results and discussion

Thermal responses of the metal thin films are strongly dependent on the pulse duration time and can be viewed with respect to the relative differences among characteristic time scales. Those time scales are (1) pulse laser duration, t_p, (2) electron relaxation time, τ_F, (3) electron-lattice thermalization time, $t_c = C_e/G$. Comparison with the values appeared in Table 1 results in

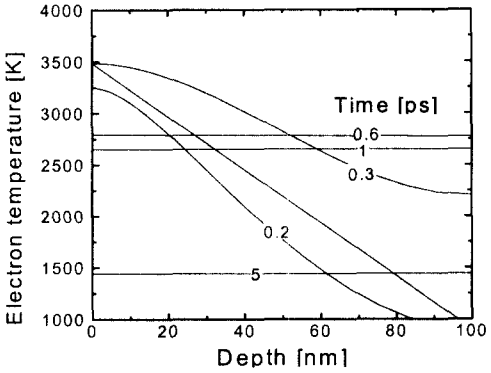
$$t_c > t_p > \tau_F \tag{15}$$

which tells that two-step method is appropriate in this study, rather than one-step method, since thermalization time is considerably longer than pulse duration.

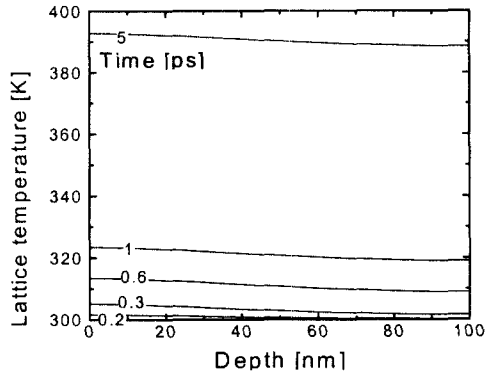
With the assumption of a very short electron relaxation time the parabolic model is appropriate to describe the heating processes, which are shown in Fig. 2. Fig. 2(a) shows the temperature distributions with respect to time, using the parabolic one step (POS) model, which assumes that the electron-lattice coupling factor is very large compared to pulse duration time. As time goes by, the lattice temperature at the front region vary according to the laser energy absorption given by Eq. (6), but the temperatures in the backside region do not change much. Fig. 2(b) and (c) depict temperature distributions of electrons and lattices calculated using the parabolic two-step (PTS) model. In Fig. 2(b) the electron temperatures in the front region increase rapidly compared to those in the rear region during the early stage of heating, while in the later time the electron



(a) Parabolic one-step model



(b) Parabolic two-step model



(c) Parabolic two-step model

Fig. 2 Temperature profiles in a 100 nm thick gold film during 0.1 ps laser pulse heating (J=500 J/m^2).

temperatures become uniform throughout the film due to the high conductivity of electrons. In Fig. 2(c), the lattice temperatures do not

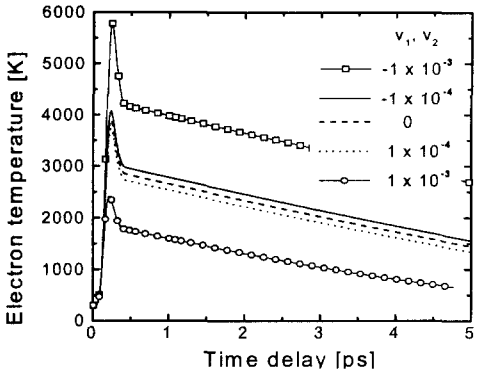


Fig. 3 Comparison of temperature profiles using HTS model for various coefficients of the complex dielectric constant.

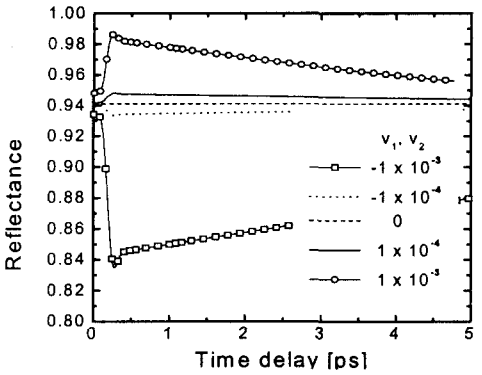


Fig. 4 Comparison of reflectances at $x=0$ using HTS model for various coefficients of the complex dielectric constant.

increase much compared to the results from POS model and remain uniformly. This implies that the metal coatings for such ultrashort pulse laser beams may not experience the high temperatures which are expected from the coatings for longer pulse laser beams.

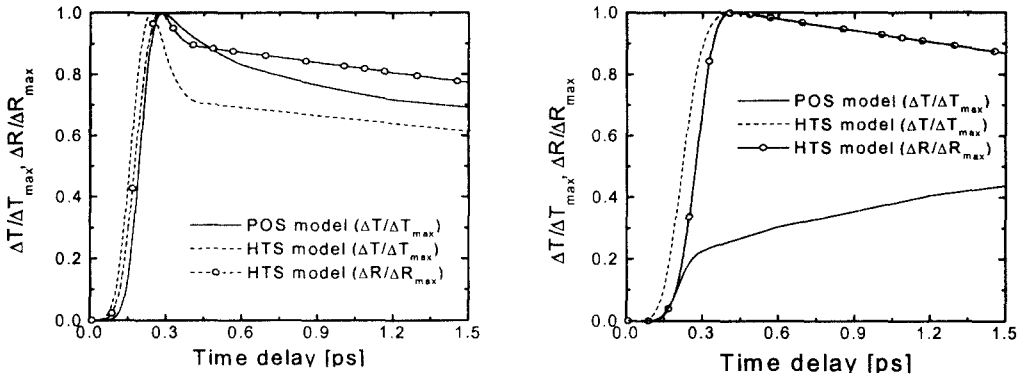
From the high electron temperatures described in Fig. 2(b), it is expected that the radiative properties can vary according to the significant change in the electron temperatures. In general, this effect has been neglected. Since high reflection mirrors have reflectances greater than 0.9, the relative change in reflectances seems to be small. That in absorptances ($A=1-R-T$), however, may not be small any more, since absorptance is smaller than 0.1 in this case. Therefore, the changed reflectance results in different temperature distributions from those based on the constant reflectance assumption.

Fig. 3 shows the temporal distributions of the maximum temperatures in the film with respect to various temperature coefficients, v_1 and v_2 in Eq. (7). Since their exact values are not available at this time, v_1 is assume to be equal to v_2. The negative value of the coefficients results in the decrease of reflectances and increase of film temperature, and vice versa. In addition, the reflectances at the front surface are given in Fig. 4 for various values of the temperature coefficients.

The two-step model is based on the finiteness of the electron-lattice thermalizaton time, which was evidenced by the measurement of reflectances. One or two hundred femtoseconds after the incidence of the laser beam, electrons arrive at the thermodynamic equilibrium and the concept 'electron temperature' becomes meaningful. Conventional studies are based on the fact that the change in electron temperatures is proportional to that in reflectances,[4,15-16] as,

$$\frac{\Delta T_e}{(\Delta T_e)_{\max}} \cong \frac{\Delta R_e}{(\Delta R_e)_{\max}} \tag{16}$$

where the subscript 'max' is for the maximum change in the properties during the heating process. Fig. 5(a) and (b) show the changes in the temperatures and reflectances at the front ($x=0$) and the rear surfaces ($x=L$). While all the changes at the front surfaces predicted from POS and HTS models have similar trends with respect to time, those at the rear surface are totally different from each other. The changes predicted from the POS model increase monotonously, while those from the HTS model have peaks around 0.4 ps just after the peak of the change at the front surface due to the



(a) $x=0$ (b) $x=L$

Fig. 5 Thermal responses and reflectances of single layer metal films during 0.1 ps laser pulse heating.

large conductivity of hot electrons. This behavior has been explored in recent experiments.[3 4,17]

The changes in the temperatures and reflectances at the front surface are compared in Fig. 6 for various values of temperature coefficients for dielectric constants. Comparison with experimental results[3] (open diamond symbols) for reflectance changes implies the optimal values for the temperature coefficients are $v_1=0$, $v_2=-5\times10^{-4}\,\mathrm{K}^{-1}$ (solid square symbols), although the experimental decay just after the peak point around 0.25 ps could be described properly by the case of $v_1=v_2=-1\times10^{-4}\,\mathrm{K}^{-1}$ (solid circle symbols). The coefficients $v_1=v_2=0$ imply that the reflectance is assumed as a constant and the change in reflectances can be estimated by the relationship of Eq. (16), as done by Qiu and Tien.[18]

Fig. 7 compares the changes in temperatures and reflectances at the rear surface. For several values of temperature coefficients the reflectances predicted from the theoretical model follow the behavior of those from experiments[3] with minor differences.

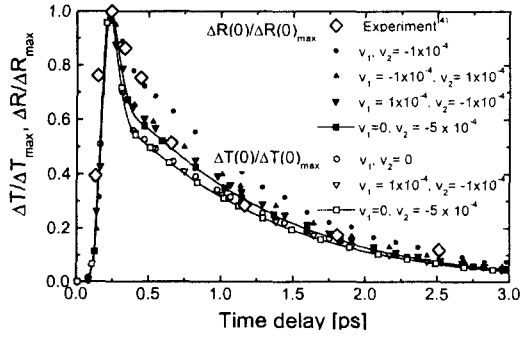


Fig. 6 Thermal responses and reflectances at $x=0$ for various coefficients of the complex dielectric constant during 96 fs laser pulse heating ($J=10\ \mathrm{J/m^2}$).

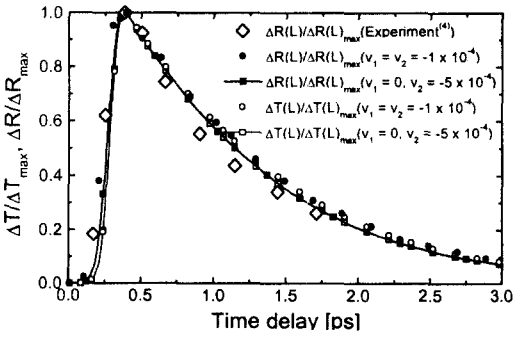


Fig. 7 Thermal responses and reflectances at $x=L$ during 96 fs laser pulse heating ($J=10\ \mathrm{J/m^2}$).

4. Conclusions

Energy transport phenomena in the metal (gold) thin-film exposed by an ultrashort pulse laser beam have been investigated by the hyperbolic two-step method.

Since the electron relaxation time is considerably shorter than the pulse duration in this study, the results from PTS and HTS models are very close to each other. The results from one-step and two step models, however, are significantly different from each other owing to the slow electron-lattice thermalization process.

Conventional approach assumes constant reflectances at the film surfaces and estimates the change in reflectances indirectly from a simple temperature-reflectance relationship. In this study, on the other hand, it is shown that the reflectances can be obtained directly from the change in dielectric constants which have a simple functional relationship with electron temperatures.

Although maximum electron temperatures in the thin-film vary within 10% for actual values of the temperature coefficients, the variation of maximum temperatures might be significant for larger values of temperature coefficients or slower thermalization processes.

For various values of temperature coefficients changes in reflectances at the front surface are different considerably, while those at the rear surface are very close to each other.

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