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TRANSIENT OPTICAL AND THERMAL RESPONSE OF METAL FILMS INDUCED BY ULTRASHORT-PULSED LASERS

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ABSTRACT

In this paper, a critical point model with three Lorentzian terms for interband transition was proposed for dielectric permittivity of metal films. After validated, it was incorporated into a two-temperature model (TTM) to study transient optical and thermal response for a copper film irradiated by an ultrashort laser pulse. The dynamic changes of reflectivity (R) and absorptivity coefficient (α) during laser irradiation, electron and lattice temperature, and phase change were investigated. It was shown that for an ultrashort laser pulse with relatively high laser fluence, both R and α could drastically decrease, leading to significantly different thermal response than that described by using constant R and α at room temperature (RT).

INTRODUCTION

Besides models themselves, thermophysical and optical properties are the key factors that govern solution accuracy in theoretical study of ultrafast laser-material interactions. The former controls thermal transport and temperature distributions in a heated medium, while the latter dictates laser energy deposition. The dynamic change in R and α due to temperature could alter laser energy deposition, both in magnitude and spatial distribution. As a result, the resulting thermal responses could be different than those described by using constant R and α at RT.

In this paper, a critical point model with three Lorentzian terms for interband transition is proposed for dielectric permittivity of metal films. After validated, it is incorporated into a TTM to study the transient optical and thermal response for a copper film irradiated by an ultrashort laser pulse. The dynamic changes of R and α during laser irradiation, distribution of laser heat density, electron and lattice temperature, and solid-liquid phase change are investigated.

MODEL DESCRIPTION

The critical model model with three Lorentzian terms is in expressed as:

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_D^2}{\omega^2 + i\gamma\omega} + \sum_{p=1}^{n} B_p \Omega_p \left(\frac{e^{i\phi_p}}{\Omega_p - \omega - i\Gamma_p} + \frac{e^{-i\phi_p}}{\Omega_p + \omega + i\Gamma_p} \right) = \varepsilon_1(x,t) + i\varepsilon_2(x,t) \quad (1)$$

where ε_∞ is dielectric constant, ω_D plasma frequency, ω laser frequency, γ damping coefficient which equals reciprocal of electron relaxation time (τ_e), n number of oscillators, B a weighting factor, and Ω, ϕ and Γ energy of gap, phase and broadening, respectively. The parameters in Eq. (1) for copper with n = 3 were best fitted experimentally with τ_e = 10.0 fs using a simulated annealing method [1]. The optical properties R and α can be determined from Fresnel function with the normal refractive index and extinction coefficient as functions of ε_1 and ε_2 [2].

Assume that a laser pulse is impinged on the front surface (x = 0). The one-dimensional TTM [1] is given as follows:

$$C_e \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x}\left(k_e \frac{\partial T_e}{\partial x}\right) - G(T_e - T_l) + S(x,t) \quad (2)$$

$$C_l \frac{\partial T_l}{\partial t} = \frac{\partial}{\partial x}\left(k_l \frac{\partial T_l}{\partial x}\right) + G(T_e - T_l) \quad (3)$$

with the laser heat density

$$S(x,t) = 0.94 \frac{[1 - R(0,t)]J_o}{t_p} \alpha \exp\left[-\int_0^x \alpha(x,t)dx - 2.77\left(\frac{t}{t_p}\right)^2 \right] \quad (4)$$

where T is temperature, C heat capacity, k thermal conductivity, G electron-phonon coupling factor, J_o laser fluence, and t_p pulse duration as FWHM. The subscript e and l denote electron and lattice, respectively. The analytical expressions of temperature-

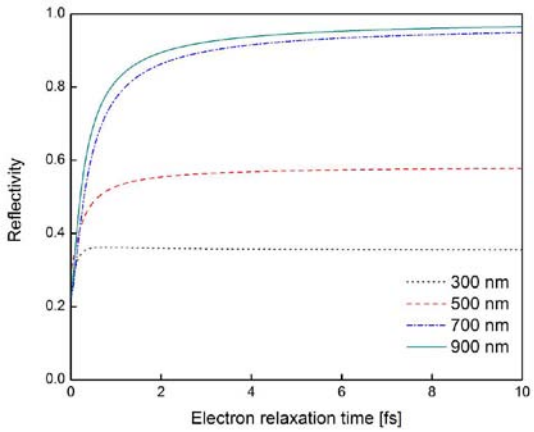

Figure 1 Reflectivity as a function of τ_e for four different laser wavelengths

dependent heat capacity, thermal conductivity and coupling factor can be found in Ref. [1].

The solid-liquid (S-L) phase change induced by ultrashort laser pulse is controlled by nucleation dynamics instead of energy balance. The S-L interface can be superheated well above the normal melting point during the melting process and undercooled far below the melting point in the solidification process. The numerical description for modeling rapid S-L phase change can be found in Ref. [4].

RESULTS AND DISCUSSION

The numerical simulations are performed for a copper film of 1-μm thickness irradiated by an ultrashort laser pulse. The initial temperature is set at 300 K.

Figures 1 and 2 show the reflectivity (R) and absorptivity coefficient (α) of copper as functions of electron relaxation time (τ_e) for four different laser wavelengths (λ), where τ_e = 0.01 fs corresponds to very high temperature and 10 fs to RT. It appears that R decreases significantly when τ_e < 3 fs, and α does when τ_e < 2 fs. The corresponding electron temperature for τ_e = 3 fs is 1.4×10^4 K based on T_l = 750 K. These temperatures can be induced in a copper film by a 100-fs, 700-nm laser pulse at fluence 0.8 J/cm^2.

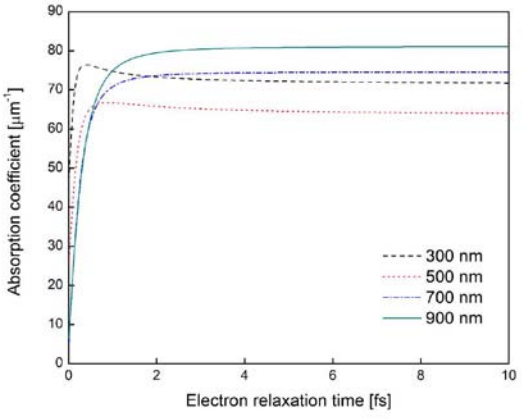

Figure 2 Absorption coefficient as a function of τ_e for four different laser wavelengths

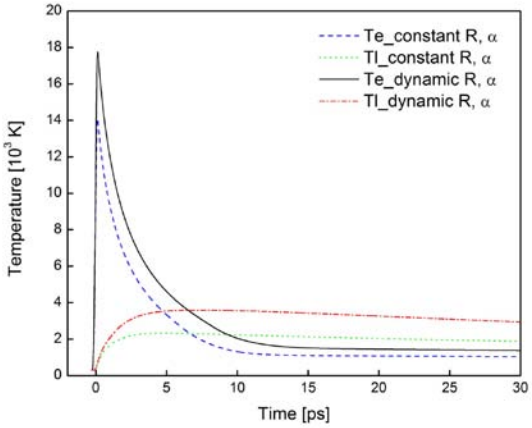

Figure 3 Electron and lattice temperature at the front surface: t_p = 150 fs, λ = 800 nm, and J_o = 1.2 J/cm^2

Figure 3 shows the time evolutions of electron and lattice temperature at the front surface of the copper film heated by a 150-fs laser pulse with J_o = 1.2 J/cm^2. Both T_e and T_l predicted with dynamic R and α are higher than those predicted with constant R and α at RT. The simulated peak T_e and T_l are 17,760 K and 3,589 K versus 14,096 K and 2,329 K, respectively. It can be seen in Fig. 3 that T_e becomes equal to T_l at about t = 6.5 ps. After that, T_e is even lower than T_l. The difference keeps increasing until t = 14.8 ps and then decreasing until the thermal equilibrium is established at about 2.5 ns. The non-equilibrium between T_e and T_l during this stage is governed by two competing mechanisms: diffusion of electron thermal energy into the deeper part of electrons and energy exchange between electrons and lattice. Obviously, the former is predominating in this case. The difference between T_e and T_l depends on laser fluence. The high the flunce is, the larger the difference is.

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