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Heat Transfer Mechanisms During Short-Duration Laser Heating of Thin Metal Films

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A perturbation technique is used to simplify the generalized governing equations of the parabolic two-step model. The generalized form of the two-step model contains diffusion terms in both electron and lattice energy equations and assumes that incident laser radiation is absorbed by both electron gas and solid lattice to account for the thermal behavior of semiconducting and impure materials. The simplified perturbation technique is used to eliminate the coupling between the electron and the lattice energy equations when the temperature difference between the electron and the lattice is a small perturbed quantity, which is true in materials exhibiting high coupling factors. A mathematical criterion is derived to determine the conditions under which electron and lattice are in thermal equilibrium. It is found that five dimensionless parameters control the state of thermal equilibrium between the lattice and the electron.

KEY WORDS: laser heating; nonequilibrium heating; pulse heating; thermal waves; thin metal films.

1. INTRODUCTION

Energy transport during fast laser heating of solids has become a very active research area due to the tremendous applications of short-pulse lasers in the fabrication of microstructures, synthesis of advanced materials, measurement of thin-film properties, diagnostics of material's structure transformation, micromachining, laser patterning, laser processing of diamond films from carbon ion-implanted copper substrates, and laser surface hardening [1, 2].

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There exist basically four models describing the mechanism of energy transport during short-pulse laser heating. The first is the parabolic one-step model, which is based on the classical Fourier conduction law. The second is the hyperbolic one-step model [3, 4], which was first postulated for gases by Maxwell [5]. The third and fourth models are the parabolic two-step and the hyperbolic two-step models [2, 6, 7].

The first two models neglect the microscopic energy deposition process and use empirical laws to describe the energy transport in materials. The microscopic mechanisms of energy deposition become important when the heating process is very fast. Laser heating of metals consists of two major steps of microscopic energy transfer. Electrons in metals first absorb radiation energy and then transmit energy to the lattice through inelastic electron–phonon scattering processes. Depending on the value of the electron–phonon coupling factor, it takes about 0.1–1 ps for electrons and lattice to reach thermal equilibrium for typical metals. When the laser pulse duration is comparable to or less than this thermalization time, the electron and lattice are not in thermal equilibrium. In these situations, it is important to use the microscopic two-step models.

The microscopic two-step model pioneered by Anisimov et al. [8] and advanced later by Fujimoto et al. [9]. The two-step model involves two coupled energy equations governing the heat transfer in the electron gas and the metal lattice. Many different assumptions limit the generality of the two-step models. These assumptions are that (a) electron–phonon interaction is the dominant scattering process for electrons, (b) the conduction of heat by phonons is negligible, and (c) the incident laser energy is totally absorbed by electron gas. In addition to the above limitations, it is not easy to solve the coupled energy equations even after eliminating the coupling between them. The elimination of the coupling between the two equations yields a single equation containing a higher-order mixed derivative in both time and space. The existence of such terms complicates the solution methodology.

Now, return to the above three assumptions, especially the second and the third assumptions, which limit the validity of the two-step model. It is known that energy deposits into materials in different ways, depending on the nature of heating methods and the structure of materials. For example, it can deposit simultaneously on all energy carriers (e.g., electrons and phonons) through contact heating at surfaces or selectively on a particular group of carriers by radiation heating. Radiation heating excites free/bound electrons in metals but excites valence electrons or optical phonons in semiconductors. As a result, each of the energy equations of the two-step models must contain a source term to account for that part of incident radiation absorbed by its energy carriers which are electrons or phonons.

Also, assumption (b) is not justified for metals containing large amount of impurities or for semiconducting materials, where it is known that energy may be diffused by both solid lattice and electron gas in these materials.

The electron–phonon coupling factor varies within a wide range of limits. Many metals have very low coupling factor. As an example, the coupling factor of gold is about $2.6 \times 10^{16}\ \mathrm{W \cdot m^{-3} \cdot K^{-1}}$. However, many other metals have a very high coupling factor. As an example, the coupling factor of vanadium is about $648 \times 10^{16}\ \mathrm{W \cdot m^{-3} \cdot K^{-1}}$. As the value of the coupling factor increases, the thermalization time, which is the time required for the electron and lattice to reach the thermal equilibrium state, decreases. This implies that the normalized temperature difference between the electron gas and the lattice becomes small but not negligible. This small temperature difference between the electron and the lattice is observed especially in materials having large coupling factors and when the laser duration time is long enough to enable the electron gas to give part of its energy to the solid lattice. When the temperature difference between the electron gas and the lattice becomes small enough, then this difference may be normalized in the form of a perturbed quantity. As a result, perturbation technique may be used to eliminate the coupling between the two energy equations. The elimination of this coupling produces two uncoupled partial differential equations which have the same order as the original coupled partial differential equations and which do not contain any mixed derivative terms.

The aim of the present work is to present a simplified perturbation technique to reformulate the generalized governing equations of the parabolic two-step model. The generalized equations contain diffusion terms in both electron gas and solid lattice energy equations to account for the thermal behavior of semiconducting materials or impure metals. Also, the generalized governing equations assume that incident laser heating is absorbed by both electron gas and solid lattice. As a study case, we consider a simple problem which is solved analytically using the proposed perturbation technique. In addition, a mathematical criterion which determine the parameters effects the state of thermal equilibrium between the electron and lattice is derived.

2. ANALYSIS

Consider applications involving short-pulse laser heating on metals. When the laser pulse duration is much shorter than the electron–phonon thermal relaxation time, the hot electrons do not have enough time to establish local thermal equilibrium with the lattice. Consequently, the electrons and the lattice have two different temperatures T_e and T_l. This

nonequilibrium heating process can be modeled phenomenologically using the two-step model as

$$C_e(T_e)\frac{\partial T_e}{\partial t} = \nabla \cdot (K_e \nabla T_e) - G(T_e - T_l) + FQ \tag{1}$$

$$C_l(T_l)\frac{\partial T_l}{\partial t} = \nabla \cdot (K_l \nabla T_l) + G(T_e - T_l) + (1 - F)\, Q \tag{2}$$

where G is the coupling factor which characterizes the energy exchange between phonon and electrons and is given as [1, 2]

$$G = \frac{\pi^4 (n_e v_s k)^2}{K_e} \tag{3}$$

$$v_s = \frac{k}{2\pi h}(6\pi^2 n_a)^{-1/3} \tag{4}$$

Equations (1) and (2) represent the generalized form of the two-step model. This generalized form assumes that a fracton F of the incident energy Q is absorbed by the electron gas and the rest $(1 - F)$ of the incident energy is absorbed by the lattice. In addition, the generalized model assumes that the diffused heat flux is carried by both free electrons and by the lattice. The inclusion of both the heating source term $(1 - F)\, Q$ and the diffusion term $\nabla \cdot (K_l \nabla T_l)$ into the energy equation of the lattice is important especially for applications involving the interaction of laser heating with metals containing a large amount of impurities, for some very particular metals, such as bismuth, or for semiconductors [10].

For pure metals, the incident radiation and the diffused heat flux are absorbed and diffused mainly by electrons, and as a result, Eqs. (1) and (2) are reduced to

$$C_e(T_e)\frac{\partial T_e}{\partial t} = \nabla \cdot (K_e \nabla T_e) - G(T_e - T_l) + Q \tag{5}$$

$$C_l(T_l)\frac{\partial T_l}{\partial t} = G(T_e - T_l) \tag{6}$$

Laser heating of thin films may be modeled as a one-dimensional problem (Fig. 1), since the beam diameter is much larger than the heat penetration depth. In addition, the thermal properties C_e, C_l, K_e, and K_l are assumed to be constant. This assumption will not restrict the generality or the validity of the solution methodology proposed in the following analysis.

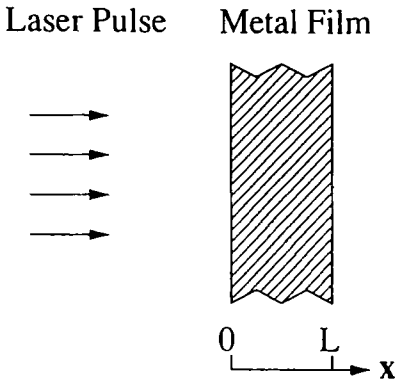

Fig. 1. Schematic diagram for laser heating of a metal film.

Now, using the dimensionless parameters defined in the nomenclature, Eqs. (1) and (2) in their one-dimensional form may be rewritten as

$$\frac{\partial \theta_e}{\partial \tau} = \frac{\partial^2 \theta_e}{\partial \xi^2} - H_1(\theta_e - \theta_l) + FP_e(\tau, \xi) \tag{7}$$

$$\frac{\partial \theta_l}{\partial \tau} = H_2 \frac{\partial^2 \theta_l}{\partial \xi^2} + H_3(\theta_e - \theta_l) + (1 - F) P_l(\tau, \xi) \tag{8}$$

where

$$H_1 = \frac{GL^2}{K_e}, \qquad H_2 = \frac{K_l C_e}{K_e C_l}, \qquad H_3 = \frac{GL^2 C_e}{K_e C_l}, \qquad P_e = P_l = \frac{L^2 Q(\tau, \xi)}{K_e T_i}$$

Equations (7) and (8) are two coupled partial differential equations which are second order in space and first order in time. Elimination of the coupling between these equations yields a mixed derivative partial differential equation which is fourth order in space and second order in time. The higher-order and mixed derivative terms that appear in the resulting equations raise the difficulty of solving such problems. However, in many applications, the coupling between the two energy equations, (7) and (8), may be eliminated without raising the order of the resulting partial differential equations and without the appearance of mixed derivative terms. These applications involve situations in which the incident thermal radiation interacts with materials having a very large coupling factor or situations in which

the laser pulse duration is not too short. In these situations, the difference between the electron and the lattice temperature may be normalized in the form of a very small perturbed quantity. This difference may be written as

$$\theta_e(\tau, \xi) = \theta_l(\tau, \xi) + \varepsilon \Delta(\tau, \xi) \tag{9}$$

where $\Delta(\tau, \xi)$ is a function of space and time, and $\varepsilon = 1/H_1$ is a dimensionless small parameter.

Consider, for example, the interaction of a laser beam with a very thin lead film of 1×10^{-6}-m thickness. For lead, the thermal conductivity and coupling factor have values of $35\ \mathrm{W \cdot m^{-1} \cdot K^{-1}}$ and $12.4 \times 10^{16}\ \mathrm{W \cdot m^{-3} \cdot K^{-1}}$, respectively [1, 7]. Under these conditions, $H_1 = 3500$, and as a result, $\varepsilon (= 1/H_1)$ may be considered a very small perturbed quantity. Examples of other metals having a very large coupling factor (i.e., having a very small perturbed parameter ε) are vanadium, niobium, and titanium.

Now Eqs. (7) and (8) can be written as

$$\frac{\partial \theta_e}{\partial \tau} = H_4 \frac{\partial^2 \theta_e}{\partial \xi^2} + C_R P_e(\tau, \xi) \tag{10}$$

$$\Delta(\tau, \xi) = -\frac{\partial \theta_e}{\partial \tau} + \frac{\partial^2 \theta_e}{\partial \xi^2} + F P_e(\tau, \xi) \tag{11}$$

where

$$H_4 = \frac{C_e}{C_e + C_l} \frac{K_e + K_l}{K_e}, \qquad C_R = \frac{C_e}{C_e + C_l}$$

Equation (10) is obtained by combining Eqs. (7) and (8), and Eq. (11) is Eq. (7) with regard to Eq. (9). It is clear that Eq. (10) is a simple partial differential equation which has the same order as that of the original governing Eqs. (7) and (8) and which has no mixed derivative terms. It is worth mentioning that the previous analysis is valid even if the original governing equations, (1) and (2), contain temperature-dependent thermal properties.

3. CASE STUDY

Consider the interaction of a short-pulse laser beam with a pure metal film having high coupling factor G. The metal film, which is of thickness L, is analyzed on the basis of a one-dimensional model, since the beam diameter is typically much larger than the heat diffusion penetration depth

in a very short time. By neglecting the temperature dependence of thermal properties, the governing equations are Eqs. (10) and (11). The heating source term in these equations is given in dimensionless form as [2, 6, 7]

$$P_e(\tau, \xi) = \frac{L^2}{K_e T_i} (1 - R)\, I\alpha \exp^{-\alpha L \xi} \exp^{-11.08} \exp^{-2.77\eta^2} \exp^{11.08\eta} \tag{12}$$

where $\eta = \tau/\tau_p$ and τ_p is the dimensionless pulse duration time. In Eq. (12) we have made a phase shift in time in the form $\tau = \hat{t} + 2$. This shift is done to enable us to take the initial conditions at time 0. Without this phase shift in time, researchers usually take the initial conditions as $\hat{t} = -2$. As a result of this phase shift, the initial and boundary conditions of the problem are given as

$$\theta_e(0, \xi) = \theta_l(0, \xi) = 0 \tag{13}$$

$$\frac{\partial \theta_l(\tau, 0)}{\partial \xi} = \frac{\partial \theta_l(\tau, 1)}{\partial \xi} = \frac{\partial \theta_e(\tau, 0)}{\partial \xi} = \frac{\partial \theta_e(\tau, 1)}{\partial \xi} = 0 \tag{14}$$

The solution of Eq. (10) is obtained using Green's function method. This solution assumes the form [11]

$$\theta_e(\tau, \xi) = C_R \int_{\tau^*=0}^{\tau} \int_{\xi^*=0}^{1} \sum_{m=0}^{\infty} \frac{1}{N(\beta_m)} \exp^{-H_4 \beta_m^2 (\tau - \tau^*)} \times \cos(\beta_m \xi) \cos(\beta_m \xi^*)\, P_e(\tau^*, \xi^*)\, d\xi^*\, d\tau^* \tag{15}$$

where

$$\beta_m = m\pi, \qquad N(\beta_m) = 0.5, \qquad N(0) = 1$$

Inserting the heating source term from Eq. (12) into Eq. (15) and carrying out the required manipulation yield

$$\theta_e(\tau, \xi) = A \sum_{m=0}^{\infty} \frac{1}{N(\beta_m)} \exp^{-H_4 \beta_m^2 \tau} \cos(\beta_m \xi)\, D_m \sqrt{\pi\delta} \times \exp^{\gamma^2 \delta} \left[\operatorname{erf}\left(\gamma \sqrt{\delta} + \frac{\tau}{2\sqrt{\delta}} \right) - \operatorname{erf}(\gamma \sqrt{\delta}) \right] \tag{16}$$

where

$$D_m = \frac{L\alpha[1-\exp^{-\alpha L}\cos(m\pi)]}{\alpha^2 L^2 + m^2\pi^2}, \qquad A = \frac{C_R L^2}{K_e T_i}(1-R)\, I\alpha$$

$$\gamma = -H_4 m^2\pi^2 - \frac{11.08}{\tau_p}, \qquad \delta = \frac{\tau_p^2}{11.08}$$

Expression for $\theta_1(\tau, \xi)$ is obtained from Eq. (9), where $\Delta(\tau, \xi)$ is obtained from Eqs. (11) and (16) as

$$\begin{aligned}\Delta(\tau,\xi) = {} & A(H_4-1)\sum_{m=0}^{\infty}\frac{1}{N(\beta_m)}(m^2\pi^2)\exp^{-H_4\beta_m^2\tau}\cos(\beta_m\xi)\, D_m\sqrt{\pi\delta} \\ & \times\exp^{\gamma^2\delta}\left[\operatorname{erf}\left(\gamma\sqrt{\delta}+\frac{\tau}{2\sqrt{\delta}}\right)-\operatorname{erf}(\gamma\sqrt{\delta})\right] \\ & -A\sum_{m=0}^{\infty}\frac{1}{N(\beta_m)}\exp^{-H_4\beta_m^2\tau}\cos(\beta_m\xi)\, D_m \\ & \times\exp^{\gamma^2\delta}\exp^{-[\gamma\sqrt{\delta}+\tau/2\sqrt{\delta}]^2}+P_c(\tau,\xi) \end{aligned} \tag{17}$$

In the previous solution, it is assumed that the incident laser radiation is totally absorbed by the electron gas, which is the case for pure metal films,

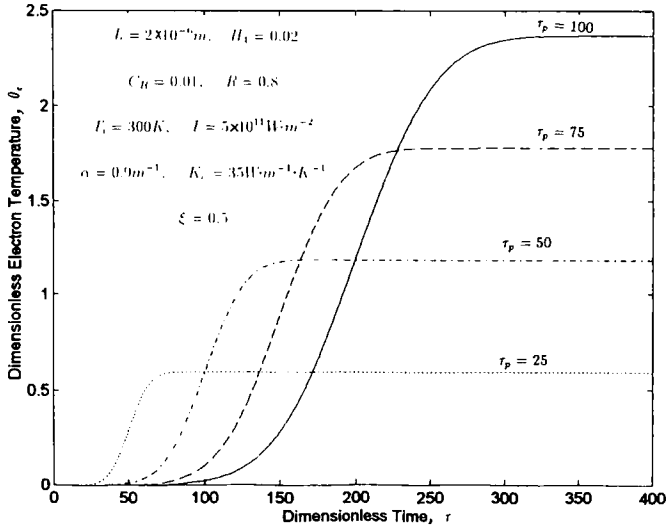

Fig. 2. The dynamic response of the electron temperature θ_e at different duration times.

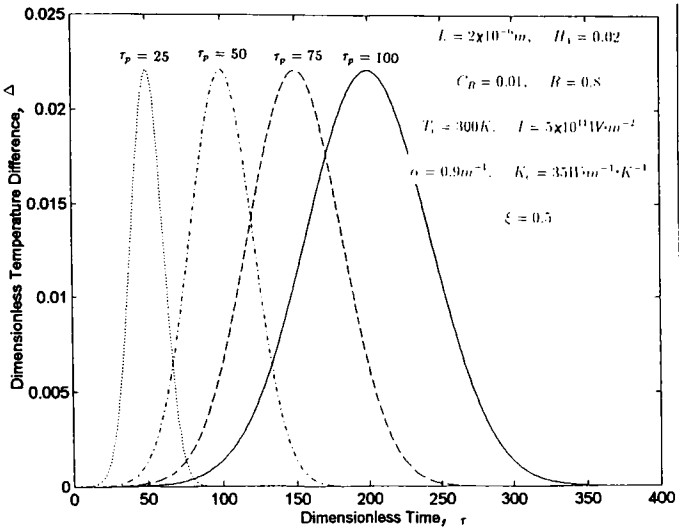

Fig. 3. The dynamic response of the temperature difference function Δ at different duration times.

and as a result $F = 1$ in Eq. (11). A sample of the results is plotted in Figs. 2 and 3, which show the dynamic response of both the dimensionless electron temperature and the dimensionless temperature difference Δ at a given location of the metal film.

4. THERMAL EQUILIBRIUM CRITERION

In the following section, a mathematical criterion is derived to determine the conditions under which both the electron gas and the lattice are in thermal equilibrium.

Let us assume that electron and lattice are in thermal equilibrium if the maximum temperature difference $(\theta_e - \theta_l)_{max}$ is less than a certain dimensionless difference q. As a result,

$$\Delta \leqslant \eta H_l \tag{18}$$

Substituting for Δ from Eq. (17) yields the required general criterion. However, it can be shown that the difference function Δ has its maximum absolute value Δ_{max} at $\xi = 0$ and $\tau = 2\tau_p$. Also, it is clear from Fig. 2 that τ_p has a weak effect on Δ_{max}. As a result, the thermal equilibrium criterion given in Eq. (18) is written as

$$A(H_4-1)\sum_{m=0}^{\infty}\frac{1}{N(\beta_m)}(m^2\pi^2)\exp^{-2H_4\beta_m^2\tau_p D_m\sqrt{\pi\delta}}$$
$$\times\exp^{\gamma^2\delta}\left[\operatorname{erf}\left(\gamma\sqrt{\delta}+\frac{\tau_p}{\sqrt{\delta}}\right)-\operatorname{erf}(\gamma\sqrt{\delta})\right]$$
$$-A\sum_{m=0}^{\infty}\frac{1}{N(\beta_m)}\exp^{-2H_4\beta_m^2\tau_p D_m}$$
$$\times\exp^{\gamma^2\delta}\exp^{-[\gamma\sqrt{\delta}+\tau_p/\sqrt{\delta}]^2}+\frac{A}{C_R}\leqslant\eta H_1 \tag{19}$$

If the criterion expressed by Eq. (18) or (19) is satisfied, one may assume that $\theta_e\approx\theta_l$. As a result, Eqs. (7) and (8) may be combined to yield the classical energy equation, which is derived based on the classical Fourier law.

It is clear from Eq. (19) that five dimensionless parameters control the thermal equilibrium state between the electron and the lattice. These parameters are H_4, αL, C_R, H_1, and A.

5. CONCLUSION

A perturbation technique is used to simplify the generalized governing equations of the parabolic two-step model. The generalized form of the two-step model contains diffusion terms in both electron and lattice energy equations and assumes that incident laser radiation is absorbed by both electron gas and solid lattice to account for the thermal behavior of semiconducting and impure materials. The simplified perturbation technique is used to eliminate the coupling between the electron and the lattice energy equations when the temperature difference between the electron and the lattice is a small perturbed quantity, which is true in materials exhibiting high coupling factors. The elimination of this coupling produces two uncoupled partial differential equations which have the same order as the original coupled partial differential equations and which do not contain any mixed derivative terms. A mathematical criterion is derived to determine the conditions under which electron and lattice are in thermal equilibrium. It is found that five dimensionless parameters control the state of thermal equilibrium between the lattice and the electron. These parameters are expressed in terms of the dimensionless coupling factor H_1, dimensionless radiation absorption coefficient αL, heat capacity ratio C_R, product of the heat capacity and thermal conductivity ratios H_4, and dimensionless fraction of the absorped incident radiation A. The approach adopted in this work is used in metal films having a high coupling factor between the

electrons and the lattice. Examples of metal films having such a high coupling factor are vanadium, lead, and titanium.

NOMENCLATURE

C Heat capacity, $J \cdot m^{-3} \cdot K^{-1}$
F Fraction of incident laser energy absorbed by electron gas
G Electron–phonon coupling factor, $W \cdot m^{-3} \cdot K^{-1}$
h Planck constant, $J \cdot s$
I Laser intensity, $W \cdot m^{-2}$
k Boltzmann constant, $J \cdot K^{-1}$
K Thermal conductivity, $W \cdot m^{-1} \cdot K^{-1}$
L Film thickness, m
n_e Electron number density per unit volume
n_a Atomic density per unit volume
P Dimensionless source term
Q Source term, $W \cdot m^{-3}$
R Metal surface reflectivity
t Time, s
t_p Laser pulse duration, s
T Temperature, K
T_i Initial temperature of both lattice and electron gas, K
T_D Debye temperature, K
v_s Speed of sound, $m \cdot s^{-1}$
x Spatial coordinate, m

Greek Symbols

α Radiation absorption coefficient, m^{-1}
Δ Difference function
ε Dimensionless small parameter
ξ Dimensionless spatial coordinate, x/L
θ Dimensionless temperature, $(T - T_i)/T_i$
τ Dimensionless time, $tK_e/(L^2C_e)$
τ_p Dimensionless laser pulse duration time, $t_pK_e/(L^2C_e)$

Subscripts

a Atom
e Electron
i Initial
l Lattice

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