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Picosecond thermal pulses in thin metal films

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A simplified approach is introduced to describe the thermal behavior of a thin metal film exposed to picoseconds thermal pulses. The approach is based on the assumption that the metal film thermal behavior occurs in two separate stages. In the first stage, electron gas transmits its energy to the solid lattice through electron-phonon coupling and other mechanisms of energy transport are negligible. In the second stage, electron gas and solid lattice are in thermal equilibrium, the energy transfer through electron-phonon coupling is negligible, and thermal diffusion dominates. The proposed approach eliminates the coupling between the energy equations and the reduced differential equations are easier to handle. The proposed approach applies to metal films whenever $GL^2/K_e \gg 1$. *© 1999 American Institute of Physics.* [S0021-8979(99)08704-6]

I. INTRODUCTION

Energy transport during fast laser heating of solids has become an active research area beginning with the employment of short-pulse lasers in the fabrication of microstructures, laser patterning, laser processing of diamond films from carbon ion implanted copper substrates and laser surface hardening,[1,2] etc.

In the literature, there are four models that describe the mechanism of energy transport during short-pulse laser heating. In the first one, the parabolic one-step model is based on the classical Fourier conduction law. In the second one, the hyperbolic one-step model,[3–5] first postulated by Maxwell,[6] is considered. In the third and fourth ones, both the parabolic and the hyperbolic two-step models[7–10] are utilized. In the first two models, the microenergy transfer is neglected and the energy transport is described by empirical laws. The microscopic mechanisms of energy deposition becomes important when the heating process is relatively fast.

Laser heating of metals consists of two major steps of micro energy transfer which occurs simultaneously. In the first step, electrons absorb most of the incident radiation energy and the excited electron gas transmits energy to the lattice through inelastic electron-phonon scattering process. In the second step, the incident radiation absorbed by the metal film diffuses spatially within the film mainly by the electron gas. For typical metals, depending on the degree of electron-phonon coupling, it takes about 0.1–1 ps for electrons and lattice to reach thermal equilibrium. When the laser pulse duration is comparable with or less than this thermalization time, electrons and lattice are not in thermal equilibrium. Under these situations, both the parabolic and hyperbolic two-step models need to be simultaneously considered.

The microscopic two-step model pioneered by Anisimov *et al.*[11] and advanced later by Fujimoto *et al.*[12] involves two coupled energy equations governing the heat transfer in the electron gas and the metal lattice. Assumptions involved with the two-step models are: (1) electron-phonon interaction is the dominant scattering process for electrons, (2) the conduction of heat by phonons is negligible, and (3) the incident laser energy is totally absorbed by electron gas.

These assumptions limit the validity of the two-step models. It is known that energy penetrates into materials in different ways, depending on the nature of heating methods and the structure of materials. For example, energy can be simultaneously transferred to electrons and phonons through contact heating at surfaces. Radiation heating excites both free and bound electrons in metals, but excites valence electrons or phonons in semiconductors. Each one of the energy equations of the two-step models must then contain a source term to account for that part of incident radiation absorbed by electrons or phonons. Assumption (2) 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 addition to these limitations of the two-step model, it is difficult to solve the coupled energy equations even after eliminating the coupling. The elimination of the coupling yields a single equation containing higher order mixed derivatives in both time and space leading to complications in a solution procedures. However, in situations involving high electron-phonon coupling and low electron thermal conductivity, a simplification of the generalized governing equations of the parabolic two-step model is possible. This can be done by realizing that the thermal behavior occurs in two separate steps. In the first stage, which is very short, electron gas absorbs the incident radiation and transmits its energy to the metal lattice. The incident radiation raises the electron gas temperature, which in turn, causes a sharp drop in its thermal conductivity as reported for the case of most metals. This implies that the energy diffused by the electron gas may be neglected compared to that transmitted to the solid lattice during the first stage. As a result, the first stage is described by the parabolic two-step model excluding the diffusion term. The coupling

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between the energy equations of the parabolic two-step model is easily eliminated after dropping this diffusion. The reduced energy equations may then be used to obtain the nonequilibrium period, that is, the time required by both the electron and the lattice to reach the local thermal equilibrium. At the end of the first stage, both electron gas and solid lattice assume the same temperature. The second stage begins following the nonequilibrium period. In this stage, the absorbed energy starts to diffuse within the thin film. Since both electron gas and solid lattice have the same temperature, the thermal behavior of the thin film is now described by the parabolic one-step model which consists of one energy equation containing the diffusion term. The initial condition for this model is obtained from the spatial electron or lattice temperature distribution given at the end of the nonequilibrium period.

The assumption that the mechanism of heat transfer in the first stage is controlled mainly by energy transfer from the electron gas to metal lattice and without any thermal diffusion by the electron gas is justified by the following reasons: Systems of high electron-phonon coupling factor have very small nonequilibrium period. During this short period, the low thermal conductivity electrons have no opportunity to diffuse energy within the metal film. The diffusion process requires a relatively much longer period, and controls the thermal behavior of the second stage.

The objective of the present work is to introduce a simplified approach which describes the thermal behavior of a thin film exposed to picosecond thermal pulses. It is assumed that the metal film thermal behavior occurs in two successive stages. In the first stage, electron gas transmits its energy to the solid lattice, and at the end of this stage, both electron gas and solid lattice reach the state of thermal equilibrium. In the second stage, the energy transfer through electron-phonon coupling is negligible and the thermal diffusion becomes important. The proposed approach eliminates the coupling between the energy equations of both solid lattice and electron gas, and it is much easier to handle the reduced partial differential equations.

II. ANALYSIS

Consider applications involving ultrafast laser heating of metal films. When the laser pulse duration is much shorter than the electron-phonon thermal relaxation time, the hot electrons are not in local thermal equilibrium with the lattice. Consequently, one describes the thermal behavior of a metal in terms of two energy balances, one for electrons and one for phonons. Electrons are characterized by a Fermi–Dirac distribution at the electron temperature T_e and the phonons by a Bose–Einstein distribution at the lattice temperature T_l. The coupling between the two systems occurs via the electron-phonon interaction. The time evolution of the energies is given by the two-step model as follows:[1,10]

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

$$C_e(T_e)\frac{\partial T_e}{\partial t}=\nabla\cdot(K_e\nabla T_e)-G(T_e-T_l)+Q_e, \tag{2}$$

where C_l denotes the lattice heat capacity, C_e the electron heat capacity, K_l the solid lattice thermal conductivity, K_e the electron gas thermal conductivity, Q_l the incident radiation absorbed by the lattice, Q_e the incident radiation absorbed by the electron gas and G denotes the coupling factor which characterizes the energy exchange between phonons and electrons and is given as:[1,2]

$$G=\frac{\pi^4(n_e v_s k_B)^2}{K_e},\quad v_s=\frac{k_B}{2\pi h}(6\pi^2 n_a)^{-1/3}T_D, \tag{3}$$

where n_e denotes the electron number density, v_s the speed of sound, k_B the Boltzmann constant, n_a the phonon density, h the Planck constant and T_D the Debye temperature. The coupling factor relative to electron diffusion is dimensionless and explains

$$\frac{GL^2}{K_e}\sim\frac{G(T_e-T_l)}{K_e[(T_e-T_l)/L^2]}=\frac{\text{Electron}-\text{phonon energy exchange}}{\text{Net diffusion of electrons}}, \tag{4}$$

where L denotes the metal film thickness. Note also that, as dimensional groups, the upper limit of sound propagation in a quantized solid is characterized by velocity

$$v_s\sim\nu_D n_a^{-1/3}, \tag{5}$$

where ν_D being the Debye frequency. This velocity can be rearranged in terms of the quantized energy, corresponding to the Debye temperature,

$$h\nu_D=k_B T_D, \tag{6}$$

to give

$$v_s\sim T_D\frac{k_B}{h}n_a^{-1/3} \tag{7}$$

which provides a dimensional interpretation of the sound velocity given in Eq. (3). Further note that Eq. (4) rearranged in terms of Eq. (3) gives

$$\frac{GL^2}{K_e}\sim\pi^4\left(\frac{n_e v_s k_B L}{K_e}\right)^2, \tag{8}$$

where, for a given temperature, the ratio

$$\frac{n_e v_s k_B T}{K_e T/L}\sim\frac{\text{electron energy flow}}{\text{electron diffusion}}=\text{Pe} \tag{9}$$

denotes a Peclet number which is a well-known dimensionless number in the phenomena-logical thermal science literature. In terms of this number, Eq. (8) may be interpreted as

$$\left(\frac{GL^2}{K_e}\right)^{1/2}\sim\text{Pe}. \tag{10}$$

Equations (1) and (2) represent the generalized form of the two-step model. In Eqs. (1) and (2), it is assumed that the conduction of heat by phonons is included and that the incident laser energy is absorbed by both electron gas and solid lattice. The assumption that conduction of heat may be carried by phonons, as well as by electrons, is justified for metals containing a large amount of impurities or for semicon-

TABLE I. Ratio GL^2/K_e for different metal films of thickness 1×10^{-6} m.

	Pb	Ag	Cu	Au
$K_e(\mathrm{W\,m^{-1}\,K^{-1}})$	35	419	386	315
$G(\mathrm{W\,m^{-3}\,K^{-1}})$	12.4×10^{16}	2.8×10^{16}	4.8×10^{16}	2.8×10^{16}
GL^2K_e	3542	66	124	88

ducting materials, where the energy diffuses in both solid lattice and electron gas.[13] It is also known that energy may excite phonons in semiconductors. So it is necessary for each one of the energy equations of the parabolic two-step model to contain a source term to account for that part of incident radiation absorbed by electrons or phonons.

Elimination of the coupling between Eqs. (1) and (2) yields an equation with mixed derivatives. The high order terms and mixed derivatives resulting from this elimination lead to solution difficulties. However, in some cases, the coupling between the two energy equations may be eliminated without these difficulties. These cases involve very large coupling and very short pulse duration.

A simplified solution procedure is presented here for elimination the coupling between Eqs. (1) and (2). This is done by recognizing the fact that the thermal behavior of the thin metal film occurs in two separate stages. In the first stage, which has very short duration, electron gas absorbs the incident laser radiation and transmits its energy to the solid lattice. The incident radiation raises the temperatures of the electron gas and solid lattice, which in turns causes a drop in their thermal conductivities K_e and K_l. As an example, it is reported[13] that the electron gas thermal conductivity for highly pure copper drops from 2×10^4 W/mK at $T=10$ K to 5×10^2 W/mK at $T=1000$ K. Also, a very short thermal pulse leads to a very short period for the first stage. During this stage, energy diffusion in electron gas and solid lattice is negligible. Equations (1) and (2) are then reduced to

$$C_l(T_l)\frac{\partial T_l}{\partial t}=G(T_e-T_l)+Q_l, \tag{11}$$

$$C_e(T_e)\frac{\partial T_e}{\partial t}=-G(T_e-T_l)+Q_e \tag{12}$$

which are valid for $0\leqslant t\leqslant t_0$, where t_0 is the period of the first stage. Note that the diffusion becomes negligible whenever the ratio of exchanged energy through electron-phonon coupling to diffused energy through conduction, in both lattice and electron, is much larger than 1, that is,

$$\frac{GL^2}{K_e}\gg1,\quad \frac{GL^2}{K_l}\gg1.$$

For most materials, $K_e>K_l$, and only the criterion $GL^2/K_l\gg1$ needs to be satisfied. Table I shows the first criterion for different metal films of thickness 1×10^{-6} m.[10]

For the period of the first stage combine Eqs. (11) and (12) to yield

$$C_l(T_l)\frac{\partial T_l}{\partial t}+C_e(T_e)\frac{\partial T_e}{\partial t}=Q_l+Q_e=Q \tag{13}$$

subject to the initial conditions

$$T_e(0,x)=T_l(0,x)=T_i.$$

At the end of the first stage t_0, it is assumed that the difference between the electron gas temperature $T_e(t_0,x)$ and solid lattice temperature $T_l(t_0,x)$ is a small difference $\Delta T(x)$, where

$$\Delta T(x)=T_e(t_0,x)-T_l(t_0,x) \tag{14}$$

with $\Delta T/T_e\ll1$ and $\Delta/T_l\ll1$. Integration of Eq. (13) over the first stage period yields

$$\int_{T_i}^{T_e(t_0,x)-\Delta T(x)}C_l\,dT_l+\int_{T_i}^{T_e(t_0,x)}C_e\,dT_e=\int_0^{t_0}Q\,dt. \tag{15}$$

Substitute for T_l from Eq. (12) into Eq. (13), yields

$$\frac{\partial}{\partial t}\left(\frac{C_e}{G}\frac{\partial T_e}{\partial t}\right)+\left(1+\frac{C_e}{C_l}\right)\frac{\partial T_e}{\partial t}=\frac{\partial}{\partial t}\left(\frac{Q_e}{G}\right)+\frac{Q}{C_l}. \tag{16}$$

Equations (15) and (16) need to be solved at each spatial location x to obtain the first stage period t_0 and the temperature distribution $T(t_0,x)=T_0=T_e(t_0,x)$ at the end of the first stage. Note that each spatial location has different t_0, and one has to select the largest t_0 in order to secure the state of thermal equilibrium between the electron gas and the solid lattice at each spatial location of the film. It is assumed that the second stage will be activated when the temperature difference between the lattice and the electron is a very small, but not zero. Waiting for the lattice to attain the exact temperature of the electron gas, that is $\Delta T=0$, increases the duration of the first stage period because the energy exchange between electron and lattice becomes very slow at the end of the first stage due to the decrease in the temperature difference between them. As a result, thermal diffusion within the film, which is assumed to be absent during the first stage, may be activated and this violates the basic assumption that controls the thermal behavior of the first stage.

For constant thermal properties C_e, C_l and G, Eq. (16) is solved as

$$T_e(t,x)=c_1+c_2e^{-\Lambda t}+\int\frac{1}{\Lambda}\left(\frac{\dot{Q}_e}{C_e}+\frac{QG}{C_lC_e}\right)dt \\ -e^{-\Lambda t}\int\frac{1}{\Lambda}\left(\frac{\dot{Q}_e}{C_e}+\frac{QG}{C_lC_e}\right)e^{\Lambda t}dt, \tag{17}$$

where c_1 and c_2 are obtained using the following initial conditions:

$$T_e(0,x)=T_l(0,x)=T_i,$$

and $\Lambda=G/C_e(1+C_e/C_l)$. Expression for $T_l(t,x)$ is obtained directly from Eq. (12), with

$$\frac{\partial T_e}{\partial t}(t,x)=-\Lambda c_2e^{-\Lambda t}+e^{-\Lambda t}\int\left(\frac{\dot{Q}_e}{C_e}+\frac{QG}{C_lC_e}\right)e^{\Lambda t}dt.$$

Also, Eq. (15) is integrated to yield

$$T_e(t_0,x)=T_0(x)=T_i+\frac{\int Q\,dt}{C_e+C_l}+\frac{C_l}{C_l+C_e}\Delta T(x). \tag{18}$$

It is worth mentioning here that the drop in the temperature of the electron gas, which occurs at the end of the first stage, causes an increase in the electron gas thermal conductivity. As a result, the thermal behavior of the second stage is activated. The governing equation which describes the thermal behavior of the thin film during the second stage is the classical parabolic energy equation replacing Eqs. (1) and (2),

$$C(T)\frac{\partial T}{\partial t}=\nabla(K\nabla T), \quad t>t_0, \tag{19}$$

where C denotes the metal film heat capacity, K the metal film thermal conductivity and T the metal film temperature. Note that the heating source term does not appear in Eq. (19) because the duration of the incident laser radiation is less than t_0. The initial condition for Eq. (19) is obtained from the temperature distribution $T(t_0,x)=T_0(x)$ at the end of the first stage. We now proceed to an example illustrating the simple approach developed in this study.

III. ILLUSTRATIVE EXAMPLE

Consider a very short laser pulse on a pure metal film of thickness L having a high coupling factor G. The metal film is analyzed on the basis of a one-dimensional model since, in a very short time, the beam diameter is typically much larger than the penetration depth of diffusion. Neglecting the temperature dependence of thermal properties, assuming the incident radiation to be totally absorbed by the electron gas, i.e., $Q_l=0$ and $Q=Q_e$, and neglecting thermal diffusion within the solid lattice, the governing equations are Eqs. (1) and (2) with constant thermal properties C_e, C_l and K_e. The source term appears in these equations may be approximated as

$$\begin{aligned}Q(t,x)=Q_e(t,x)&=(1-R)I\alpha\exp^{-\alpha x}\,\delta(t-t^*)\\&=Q_0(x)\,\delta(t-t^*),\end{aligned} \tag{20}$$

where $Q_0(x)=(1-R)I\alpha\exp^{-\alpha x}$, I denotes the laser intensity, R the metal surface reflectivity, α the radiation absorption coefficient, x the spatial coordinate in the film transverse direction and $\delta(t-t^*)$ the Dirac delta function which assumes that the incident laser beam evolves all of its energy at time t^*.

Insert the specified heating source, given by Eq. (20), into Eqs. (17) and (18), and carry out the required integrations, yields

$$\begin{aligned}&T_e(t_0,x)=T_0(x)=T_i+Q_0(x)[B_1+B_2e^{-\Lambda(t-t^*)}],\\&\qquad\qquad \text{for } t>t^*,\\&T_e(t_0,x)=T_0(x)=0, \quad \text{for } t<t^*,\end{aligned} \tag{21}$$

where

$$B_1=\frac{1}{C_l+C_e}, \quad B_2=\frac{C_l}{C_e(C_l+C_e)},$$

$$T_e(t_0,x)=T_0(x)=T_i+\frac{Q_0(x)}{C_l+C_e}+\frac{C_l}{C_l+C_e}\Delta T(x). \tag{22}$$

The duration of the first stage t_0 is found by equating Eq. (21) to Eq. (22) and solving for $t=t_0$, to yield

$$t_0=t^*-\frac{1}{\Lambda}\ln\left[\frac{C_e\Delta T(x)}{Q_0(x)}\right]. \tag{23}$$

During the first stage, thermal diffusion is absent, and as a result, one may assume that the temperature difference $\Delta T(x)$ has the same spatial distribution as the spatial distribution of the heating source. This implies that

$$\Delta T(x)=Q_0(x)\Delta T$$

and Eq. (23) is reduced to

$$t_0=t^*-\frac{1}{\Lambda}\ln(C_e\Delta T).$$

Expression for $T_l(t,x)$ is obtained directly from Eq. (12), with

$$\frac{\partial T_e}{\partial t}(t,x)=-Q_0(x)B_2\Lambda e^{-\Lambda(t-t^*)}, \quad \text{for } t>t^*,$$

$$\frac{\partial T_e}{\partial t}(t,x)=0, \quad \text{for } t<t^*.$$

Also, the governing equation describing the thermal behavior of the thin film during the second stage and is given by Eq. (19) is reduced to

$$C\frac{\partial T}{\partial t}=K\frac{\partial^2 T}{\partial x^2}, \quad t\geqslant t_0 \tag{24}$$

which has the following initial condition given by Eq. (22), with negligible temperature difference ΔT, and the boundary conditions

$$T(t_0,x)=T_0=T_i+\frac{(1-R)I\alpha\exp^{-\alpha x}}{C_l+C_e},$$

$$\frac{\partial T}{\partial x}(t,0)=\frac{\partial T}{\partial x}(t,L)=0. \tag{25}$$

In terms of the appropriate Green's[14] function, Eqs. (24) and (25) assume the following solution:

$$\begin{aligned}T(t,x)=&\frac{2(1-R)I\alpha^2}{C_l+C_e}\sum_{m=1}^{\infty}e^{-(K/C)\beta_m^2(t-t_0)}\\&\times\frac{\cos(\beta_m x)}{L}\,\frac{[1-\cos(\beta_m L)e^{-\alpha L}]}{\alpha^2+\beta_m^2}.\end{aligned} \tag{26}$$

where $\beta_m=(2m-1)\pi/2L$. Figure 1 shows the approximate and exact transient electron and lattice temperatures variations at the surface of an insulated thin lead film exposed to a laser pulse in the form of Eq. (20). The exact results are obtained from the numerical solution of Eqs. (1) and (2) for the same specified heat source and boundary conditions and the approximate solution is that obtained from the proposed method. It is clear from the figure that the results have good qualitative and quantitative agreement. This is because lead has the highest GL^2/K_e ratio among most metals as shown in the previous table. The results are obtained using the following properties of lead and laser pulse:

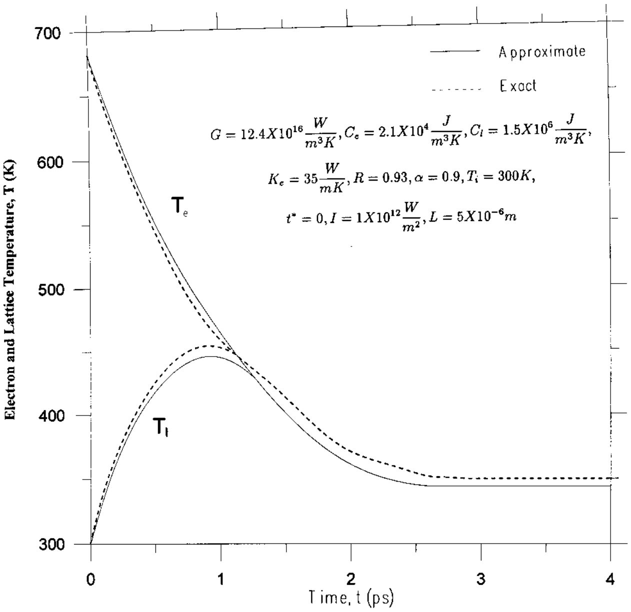

FIG. 1. Transient electron and lattice temperatures variation at the surface of an insulated thin lead film. $x=0$, $t^*=0$.

$$G=12.4\times10^{16}\ \mathrm{W/m^3\,K},\ \ C_e=2.1\times10^4\ \mathrm{J/m^3\,K},$$

$$C_l=1.5\times10^6\ \mathrm{J/m^3\,K},$$

$$K_e=35\ \mathrm{W/mK},\ \ R=0.93,\ \ \alpha=0.9,\ \ T_i=300\ \mathrm{K},$$

$$t^*=0,\ \ I=1\times10^{12}\ \mathrm{W/m^2},\ \ L=5\times10^{-6}\ \mathrm{m}.$$

IV. CONCLUDING REMARKS

A simplified approach is introduced to describe the thermal behavior of a thin film exposed to a picosecond duration thermal pulse. It is assumed that the film thermal behavior occurs in two separate stages. In the first stage, electron gas transmits its energy to the solid lattice, excluding any other mechanism of energy transport. This is believed to be true for thin, but not very thin, films having very high coupling factor, very short thermal pulse duration and relatively low or moderate thermal conductivity. As a result, thermal diffusion has not enough time to affect the film during the first stage. In the second stage, in which both electron gas and solid lattice are in thermal equilibrium, the energy transfer through electron-phonon coupling is negligible and the energy transfer by diffusion becomes dominant. The proposed approach eliminates the coupling between the energy equations of both solid lattice and electron gas and as a result, it is much easier to solve the reduced partial differential equations. The proposed approach applies on metal films satisfying the condition $GL^2/K_e \gg 1$.

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