NUMERICAL SIMULATION OF HEAT TRANSFER MECHANISMS DURING FEMTOSECOND LASER HEATING OF NANO-FILMS USING 3-D DUAL PHASE LAG MODEL

Illayathambi Kunadian, J. M. McDonough, K. A. Tagavi
Department of Mechanical Engineering
University of Kentucky
Lexington, Kentucky 40506-0503
Email: ikuna0@engr.uky.edu

ABSTRACT
In the present work we investigate femtosecond laser heating of nanoscale metal films irradiated by a pulsating laser in three dimensions using the Dual Phase Lag (DPL) model and consider laser heating at different locations on the metal film. A numerical solution based on an explicit finite-difference method has been employed to solve the DPL heat conduction equation. The stability criterion for selecting a time step size is obtained using von Neumann eigenmode analysis, and grid function convergence tests have been performed. The energy absorption rate, which is used to model femtosecond laser heating, has been modified to accommodate for the three-dimensional laser heating. We compare our results with classical diffusion and hyperbolic heat conduction models and demonstrate significant differences among these three approaches. The present research enables us to study ultrafast laser heating mechanisms of nano-films in 3D.

NOMENCLATURE

$C$ Volumetric heat capacity, $Jm^{-3}K^{-1}$
$CC$ thermal wave speed, $ms^{-1}$
$G$ Phonon-electron coupling factor, $Wm^{-3}K^{-1}$
$J$ Laser fluence, $Jm^{-2}$
$S$ Energy absorption rate, $Wm^{-3}$
$R$ Reflectivity, (dimensionless)
$T$ Temperature, $K$

$k$ Thermal conductivity, $Wm^{-1}K^{-1}$
$q$ Heat Flux density, $Wm^{-2}$
$t$ Physical time, $s$
$x$ Distance, $m$
$δ$ Laser penetration depth, $m$
$α$ Thermal diffusivity, $m^2s^{-1}$
$Δx$, $Δt$ Space step and time step
$k_B$ Boltzmann constant
$L_x$, $L_y$ Width and height of the film, $m$
$n_e$ Number density of free electrons per unit volume
$r_o$ Thickness of laser beam, $m$
$t_p$ Time, $s$
$V_s$ Speed of sound, $ms^{-1}$

INTRODUCTION
According to classical heat conduction theory heat flux is directly proportional to temperature gradient (Fourier’s law) in the form

$$-\bar{q}(\vec{r},t) = -k\nabla T(\vec{r},t)$$

(1)

When the above equation is incorporated into the first law of thermodynamics,

$$-\nabla \cdot \bar{q}(\vec{r},t) = C_p\frac{dT(\vec{r},t)}{dt}$$

(2)

*Address all correspondence to this author.
a parabolic heat conduction equation for the temperature field is obtained.

$$\nabla^2 T = \frac{1}{\alpha} \frac{\partial T}{\partial t}, \quad \text{with} \quad \alpha = \frac{k}{C_p} \quad (3)$$

Although Fourier’s law represents one of the best models in mathematical physics, it possesses anomalies, the most prominent being its prediction that heat conduction is a diffusion phenomenon in which temperature disturbances will propagate at infinite velocities. The parabolic character of Fourier’s law implies that the heat flow starts (vanishes) simultaneously with the appearance (disappearance) of a temperature gradient, thus violating the causality principle, which states that two events, which are causally correlated, cannot happen at the same time, but the cause must precede the effect, noted by Cimmelli [1]. In situations dealing with transient heat flow for extremely short periods of time (applications involving laser pulses of nanosecond and picosecond duration), high heat fluxes, and at temperatures near absolute zero (heat conduction at cryogenic temperatures), Fourier’s law fails to predict the correct temperature distribution.

In order to address this discrepancy, a modified heat flux that accommodates the finite propagation speed of observed thermal waves was proposed by Vernotte [2] and Cattaneo [3] in 1958:

$$\bar{q}(\vec{r},t) + \tau \frac{\partial \bar{q}(\vec{r},t)}{\partial t} = -k \nabla T(\vec{r},t) \quad (4)$$

where $\tau$ is the relaxation time, which is the effective mean free path divided by the phonon speed ($V_s$, speed of sound). In the absence of relaxation time ($\tau = 0$), implying infinite phonon speed or zero mean free path, Eq. (4) reduces to classical Fourier’s law. When Eq. (4) is coupled with the energy Eq. (2) we obtain hyperbolic heat conduction equation:

$$\nabla^2 T = \frac{1}{\alpha} \frac{\partial T}{\partial t} + \frac{\tau}{CC} \frac{\partial^2 T}{\partial t^2}, \quad \text{with} \quad \alpha = \frac{k}{C_p}, \quad \text{and} \quad CC = \sqrt{\frac{\alpha}{\tau}} \quad (5)$$

The above equation is the thermal wave equation depicting a temperature disturbance propagating as a wave: thermal diffusivity appearing as a damping effect in heat propagation. The quantity $CC$ is the thermal wave speed which approaches infinity when $\tau = 0$, reducing the above equation to the classical diffusion equation. The frequently cited experimental evidence for hyperbolic conduction includes that of Kaminski [4] and Mitra et al. [5], who investigated wet sand and processed meat, respectively. But later, investigations by Graßmann et al. [6] and Herwig et al. [7] clearly showed that the hyperbolic effect does not appear in their experiments for the materials for which contrary evidence was published by Kaminski and Mitra et al. Also, the hyperbolic heat conduction equation (HHCE) suffers from the theoretical problem of compatibility with the second law of thermodynamics. A parallel and infinitely wide slab subjected to sudden temperature change on its boundaries has been found to have temperature rise in the interior of the slab greater than the boundaries [8,9]. Barletta and Zanchini [9] argue that Clausius’ inequality implies that within the scheme of local equilibrium, the entropy production rate must be non-negative, but the temperature rise in the interior of the slab is accompanied by negative values of entropy, indicating that HHCE is incompatible with the local equilibrium scheme, which further implies no violation of the second law occurs: the temperature field cannot be interpreted in the usual thermodynamic sense. Kroner and Bergmann [10] point out that the hyperbolic approach to heat current density violates the fundamental law of energy conservation. Consequently the HHCE predicts physically impossible solutions with negative local heat content. In order to compensate for this defect, Bai and Lavine [11] modified the hyperbolic heat conduction equation by simply adding terms to the energy balance while making no attempt to eliminate the unrealistic results. To date, there has been no clear experimental evidence supporting hyperbolic heat conduction although wave nature has been observed by Peshkov [12] using superfluid liquid helium at temperature near absolute zero. He referred to this phenomenon as second sound, because of similarity between observed thermal and ordinary acoustic waves. Also, HHCE neglects the energy exchange between the electrons and the lattice, and so their applicability to short-pulse laser applications becomes questionable.

Recently, an increasing interest has developed in the use of lasers in numerous applications related to material processing (e.g., laser forming, laser cladding, laser welding, laser drilling, laser glazing etc.), scientific research (e.g., study of heat transfer behavior in microphotonic or electronic devices) and medicine (laser surgery). High power short-pulse lasers have given rise to several innovative technologies and have brought considerable attention to the energy transport processes occurring in materials during and after short-pulse laser material interaction. Ultrafast laser heating of metals from a microscopic point of view according to Qui and Tien [13], is composed of three processes: deposition of radiation energy on electrons, the transport of energy by electrons, and heating of the material lattice through electron-lattice interactions. During laser metal interactions, the laser pulse duration is not long enough compared with the thermalization time; a state of non-equilibrium is generated between the electrons and lattice. Qui and Tien [13] define thermalization time as the time required for the electrons and the lattice to reach thermal equilibrium, or the finite time required to convert the radiation energy into internal energy of the lattice. For relatively slow heating processes, the deposition of radiation energy can be assumed to be instantaneous and therefore can be modeled by Fourier conduction, but its applicability to very short-laser pulse duration becomes ques-
tionable: we must look for non-Fourier models.

Anisimov et al. [14] proposed a two-step model to describe the electron temperature $T_e$ and the lattice temperature $T_l$ during the short-pulse laser heating of metals. The equations describing the heating of electrons and heating of metal lattice are given by

$$ C_e \frac{dT_e}{dt} = \nabla \cdot \vec{q} - G(T_e - T_l) \quad (6) $$

$$ C_l \frac{dT_l}{dt} = G(T_e - T_l), \quad G = \frac{\pi^2 (n_e V k_B)^2}{k} \quad (7) $$

respectively, where $C_l$ and $C_e$ are the volumetric heat capacities of metal lattice and electron-gas, respectively; the phonon-electron coupling factor $G$ stands for the energy exchange between phonons and electrons per unit time; $V_e$ is the speed of sound; $n_e$ is the number density of free electrons per unit volume; $k_B$ is Boltzmann constant, and $k$ is thermal conductivity. Substitution of Eq. (1) into Eq. (6) results in parabolic two-step model and substitution of Eq. (4) in Eq. (6) results in hyperbolic two-step model. Combining Eqs. (6) and (7) and eliminating electron-gas temperature ($T_e$) gives

$$ \nabla^2 T_l + \frac{\alpha_e}{C_e} \frac{\partial (\nabla^2 T_l)}{\partial t} = \frac{1}{C_e} \frac{\partial^2 T_l}{\partial t^2} + \frac{1}{\alpha_e} \frac{\partial T_l}{\partial t} \quad (8) $$

whereas eliminating metal-lattice temperature ($T_l$) gives

$$ \nabla^2 T_e + \frac{\alpha_e}{C_e} \frac{\partial (\nabla^2 T_e)}{\partial t} = \frac{1}{C_e} \frac{\partial^2 T_e}{\partial t^2} + \frac{1}{\alpha_e} \frac{\partial T_e}{\partial t}, \quad (9) $$

where,

$$ \alpha_e = \frac{k}{C_e + C_l}, \quad C_e = \sqrt{\frac{kG}{C_e C_l}} \quad (10) $$

When investigating macroscopic effects a different model is required. Tzou [15,16,17] formulated a modified heat flux vector to account for macro- to microscale effects:

$$ \vec{q}(\vec{r}, t + \tau_q) = -k \nabla T(\vec{r}, t + \tau_q) $$

$$ \Rightarrow \vec{q}(\vec{r}, t) + \tau_q \frac{\partial \vec{q}(\vec{r}, t)}{\partial t} = -k \left( \nabla T(\vec{r}, t) + \tau_q \frac{\partial [\nabla T(\vec{r}, t)]}{\partial t} \right) \quad (11) $$

When heat flow arrives at a compound system of phonons and electrons at time $t$, the temperature gradient across the same volume can only be established later, at $t + \tau_T$, because it requires a finite duration $\tau_T$ to raise the temperature of the metal lattice by one degree. When heat flow leaves the compound system at time $t + \tau_q$ after another finite duration $\tau_q$ is required for effective collisions between phonons and electrons to take place for heat transport. The phase lag, $\tau_q$ refers to the finite time required to raise the temperature of the compound system by one degree. In short, $\tau_T$ indicates the delay behavior in establishing the temperature gradient and $\tau_q$ indicates the delay behavior in heat-flow departure.

Eq. (12) coupled with the equation of energy conservation (2) gives the DPL heat conduction equation

$$ \nabla^2 T + \tau_q \frac{\partial (\nabla^2 T)}{\partial t} = \frac{\tau_T}{\alpha} \frac{\partial^2 T}{\partial t^2} + \frac{1}{\alpha} \frac{\partial T}{\partial t} \quad (12) $$

We can see that the DPL Eq. (12) has exactly the same form as Eqs. (8) and (9). Comparing the coefficients of Eq. (12) with those of Eqs. (8) and (9) we can represent the microscopic properties as

$$ \alpha = \alpha_e \rightarrow \frac{k}{C_e C_l}, \quad \tau_T = \frac{\alpha_e}{C_e^2} \rightarrow \frac{C_l}{G}, \quad \tau_q = \frac{\alpha_e}{C_e^2} \rightarrow \frac{C_l C_e}{G(C_e + C_l)} \quad (13) $$

The microscopic effect vanishes when the phonon-electron coupling factor $G$ approaches infinity, implying that $\tau_q$ and $\tau_T$ becomes zero, reducing Eq. (12) to the classical parabolic heat conduction equation. $\tau_T = 0$ results in hyperbolic solution. Thus the DPL model covers a wide range of physical responses from the microscopic to macroscopic scales in both space and time under special values of relaxation times corresponding to the temperature gradient $\tau_T$ and heat flux $\tau_q$, respectively. DPL model looks very promising for future research because it shows very good agreement with experiments across a wide range of length and time scales.

In this paper, we will present numerical simulation of femtosecond pulse laser heating of nanometer sized gold film using DPL, classical and the hyperbolic models in 1-D, and compare with the experimental results by Brorson et al. [18], and Qiu et al. [13]. No energy loss is expected to occur during the picosecond transient, therefore both the front and rear surface boundaries are assumed to be thermally insulated. The energy absorption rate is used to model the femtosecond pulse laser heating. In continuing the investigation in the transient response on a nanometer sized gold film, we present three-dimensional formulation for an analogous problem. Now we look at pulsating laser beam heating the top of the gold film at various locations.
of the film every 0.3\(\text{ps}\) using DPL, hyperbolic and the classical parabolic model. The energy absorption rate is now modified to accommodate for the three-dimensional laser heating. The finite difference method for this problem is presented and von Neumann eigenmode analysis [19] for the finite difference algorithm is introduced along with three-dimensional stability and convergence criterion.

**MATHEMATICAL FORMULATION**

The DPL model represented by Eqn. (11) coupled with the equation of energy conservation with source term

\[-\nabla \cdot q + S(\vec{r}, t) = C \frac{\partial T}{\partial t}(\vec{r}, t) \quad (14)\]

gives the DPL heat conduction equation

\[\nabla^2 T + \tau \frac{\partial (\nabla^2 T)}{\partial t} + \frac{1}{k} \left( S + \frac{\partial S}{\partial t} \right) = \frac{\tau}{\alpha} \frac{\partial^2 T}{\partial t^2} + \frac{1 \partial T}{\partial t} \quad (15)\]

Femtosecond laser heating is modeled by the energy absorption rate given by

\[S(x, t) = 0.94J \left[ \frac{1 - R}{t_p \delta} \right] \exp \left( -\frac{x}{\delta} - \frac{1.992 | t - 2t_p |}{t_p} \right) \quad (16)\]

which is extended to 3D source term of the form

\[S(\vec{r}, t) = 0.94J \left[ \frac{1 - R}{t_p \delta} \right] \exp \left( -\frac{(x - L_x)^2 + (y - L_y)^2}{2r_o^2} - \frac{z}{\delta} - \frac{1.992 | t - 2t_p |}{t_p} \right) \quad (17)\]

where, \(r_o\) is thickness of laser beam; \(z\) is the laser penetration direction; \(L_x\) and \(L_y\) are the width and height of the nanofilm, respectively; \(t\) is the laser penetration depth and \(J\) is the laser fluence. Spatial distribution of the laser pulse in the \(x\) and \(y\) directions is assumed to be Gaussian, and uniform temperature distribution is assumed in the laser penetration direction \(z\). \(t_p\) describes laser heating of the electron-phonon system from a thermalization state. The temporal shape of the laser is Gaussian [13] with initial time shifted to \(-2t_p\), to reflect the full width at half maximum (FWHM) pulse duration, peaking at time zero. The constant 1.992 is determined from the normalized autocorrelation function of the 96\(\text{fs}\) laser pulse \((t_p)\) [13,18,20]. Initial and the boundary conditions are

\[T(\vec{r}, 0) = T_0, \quad \frac{\partial T}{\partial t}(\vec{r}, 0) = 0 \quad (18)\]

Heat loss from the film surface is negligible during the short heating period; therefore insulated boundary conditions are imposed on all sides of the film

\[\frac{\partial T}{\partial n} = 0 \quad \text{where,} \quad n = x, y, z \quad (19)\]

**NUMERICAL ANALYSIS**

An explicit finite-difference scheme [18] has been employed to solve equation (15). Centered differencing approximates the second-order derivatives in space:

\[\frac{\partial^2 T}{\partial x^2} = \frac{1}{\Delta x^2} [T^n_{i+1,j,l} - 2T^n_{i,j,l} + T^n_{i-1,j,l}] \quad (20)\]

\[\frac{\partial^2 T}{\partial y^2} = \frac{1}{\Delta y^2} [T^n_{i,j+1,l} - 2T^n_{i,j,l} + T^n_{i,j-1,l}] \quad (21)\]

\[\frac{\partial^2 T}{\partial z^2} = \frac{1}{\Delta z^2} [T^n_{i,j,l+1} - 2T^n_{i,j,l} + T^n_{i,j,l-1}] \quad (22)\]

The mixed derivative is approximated using a centered difference in space and backward difference in time

\[\frac{\partial^3 T}{\partial t \partial x^2} = \frac{1}{\Delta t \Delta x^2} [T^n_{i+1,j,l} + T^n_{i-1,j,l} - T^n_{i+1,j,l}] \quad (23)\]

\[\frac{\partial^3 T}{\partial t \partial y^2} = \frac{1}{\Delta t \Delta y^2} [T^n_{i,j+1,l} + T^n_{i,j-1,l} - T^n_{i,j,l}] \quad (24)\]
\[
\frac{\partial^3 T}{\partial t \partial x^2} = \frac{1}{\Delta t \Delta x^2} \left[ T_{i,j,l+1} - 2 T_{i,j,l} + T_{i,j,l-1} - T_{i,j,l-1} \right] + 2T_{i,j,l} - T_{i,j,l-1}^{-1} \tag{25}
\]

Forward differencing approximates the first-order derivative in time
\[
\frac{\partial T}{\partial t} = \frac{1}{\Delta t} \left[ T_{i,j,l+1}^n - T_{i,j,l}^n \right] \tag{26}
\]

and centered differencing approximates the second-order derivative in time
\[
\frac{\partial^2 T}{\partial t^2} = \frac{1}{(\Delta t)^2} \left[ T_{i,j,l}^{n+1} - 2T_{i,j,l}^n + T_{i,j,l}^{n-1} \right] \tag{27}
\]

Centered differencing is employed for the time derivative in the source term
\[
\frac{\partial S}{\partial t} = \frac{1}{2\Delta t} \left[ S_{i,j,l}^{n+1} - S_{i,j,l}^{n-1} \right] \tag{28}
\]

The stability criterion for the 3-D DPL model obtained using Von Neumann eigenmode analysis [18] is given by
\[
\Delta t \left( 2 \Delta t + 4 \tau_y \right) + \Delta t \left( 2 \Delta t + 4 \tau_x \right) + \Delta t \left( 2 \Delta t + 4 \tau_y \right) \leq 1 \tag{29}
\]

The maximum allowed time increment to achieve stable and convergent solutions under a prescribed space increment is then found to be
\[
\Delta t = \frac{-b \pm \sqrt{b^2 - 4ac}}{2a} \text{ where,}
\]
\[
a = -2\Delta x \Delta y \Delta z^2 + \Delta x^2 \Delta z^2 + \Delta x^2 \Delta y^2 \\
b = \Delta x^2 \Delta y^2 \Delta z^2 - 4\alpha \Delta y \Delta z^2 \tau_y - 4\alpha \Delta x \Delta z^2 \tau_x - 4\alpha \Delta y \Delta z^2 \tau_x \\
c = \Delta x^2 \Delta y^2 \Delta z^2 \tau_y \tag{30}
\]

RESULTS AND DISCUSSION

Firstly, we present 1D results of femtosecond pulse laser heating of 100nm thick gold film. Fig. 1 shows the experimental results of reflectivity change by Brorson et al. [18] and Qiu et al. [13] at the front surface of the gold film, and numerical predictions from parabolic, hyperbolic and DPL models. The energy absorption rate given by Eq. (16) is used to model laser heating in 1D. No energy loss is expected to occur during the picosecond transient, therefore both the front and rear surface boundaries are assumed to be thermally insulated. The DPL model agrees with experimental results very well compared to hyperbolic and parabolic models. In 1D and 3D calculations, we have set \( \tau_T = 0 \) and \( \tau_q = 0 \) to produce parabolic solution and \( \tau_T = 0 \) to produce hyperbolic solution.

Next, we proceed to 3-D simulations: Fig. 2 shows schematic of pulsating laser beam of 200nm diameter, heating the top of the gold film at various locations of the film every 0.3ps using DPL. The energy absorption rate given by Eq. (17) is used to model three-dimensional laser heating. We assume insulated boundary walls and constant thermal properties (thermal properties do not change much in the temperature range involved in this particular problem). To start with, a uniform grid 101 \( \times 101 \times 21 \) was used. The simulation was performed for a duration of 2.5ps. For \( \Delta x = \Delta y = \Delta z = 5nm \), \( \Delta t \) which satisfies the stability criterion was found to be 3.27ps. Furthermore, different grids 51 \( \times 51 \times 11 \) and 201 \( \times 201 \times 41 \) were used to check the consistency of the numerical solution in the form of grid function convergence tests. Fig. 2 shows the temperature plots obtained on the top surface of the gold film at \( t = 0.3ps \) using grids 51 \( \times 51 \times 11 \), 101 \( \times 101 \times 21 \), 201 \( \times 201 \times 41 \). We can see from Fig. 3 that reducing the step size by a factor of two and time step by a factor of four results in reduction of error by a factor of four, implying the validity of the numerical solution. The laser pulse is moved to a new location every \( \sim 0.3ps \). For better comparison between the three models, we have chosen temperature distribution at \( t = 1.56ps \). Fig. 4, Fig. 5 and Fig. 6 show the temperature distribution of the gold at 1.56ps, predicted by DPL, hyperbolic and parabolic heat conduction models, respectively. We can see that the hyperbolic and parabolic model show elevated temperature compared to the DPL model. In spite of the fact that we do not have experimental results to validate our 3D simulations, since DPL results agree with experimental results in 1D, we believe that it should be physically realizable in 3D.

CONCLUSION

DPL model agrees closely with experimental results in one dimension compared to the classical and the hyperbolic models. Energy absorption rate used to model femtosecond laser heating in 1-D is modified to accommodate for three-dimensional laser heating. Simulation of 3-D laser heating at various locations of thin film has been carried out using pulsating laser beam
THE FRONT SURFACE OF GOLD FILM OF 100nm THICKNESS: α = 1.2 × 10^{-4}m^2s^{-1}, k = 315Wm^{-1}K^{-1}, τ_p = 90ps, τ_q = 8.5ps.

(~ 0.3ps pulse duration) to compare different models. Stability criterion for selecting a numerical time step is obtained using von Neumann eigenmode analysis: Δx = Δy = Δz = 5nm, Δt = 3.27fs. Different grids (51 × 51 × 11, 101 × 101 × 21 and 201 × 201 × 41) were used to check convergence in numerical solution. From the results we infer that the parabolic and hyperbolic models over predict temperature distribution during ultrafast laser heating compared to DPL model. We believe that since the DPL model agrees with experimental results, its 3-D extension ought to be physically realized.

REFERENCES
Figure 2. 3-D SCHEMATIC OF LASER HEATING OF GOLD FILM AT DIFFERENT LOCATIONS

Figure 3. TEMPERATURE PLOTS OBTAINED ON THE TOP SURFACE OF THE GOLD FILM AT $t = 0.3\text{ps}$ USING GRIDS $51 \times 51 \times 11$, $101 \times 101 \times 21$, $201 \times 201 \times 41$

Figure 4. TEMPERATURE DISTRIBUTION OF GOLD FILM AT $1.56\text{ps}$ PREDICTED BY DPL MODEL

Figure 5. TEMPERATURE DISTRIBUTION OF GOLD FILM AT $1.56\text{ps}$ PREDICTED BY HYPERBOLIC MODEL

Figure 6. TEMPERATURE DISTRIBUTION OF GOLD FILM AT $1.56\text{ps}$ PREDICTED BY PARABOLIC MODEL