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NUMERICAL SIMULATION OF THERMAL PROCESSES PROCEEDING IN A MULTI-LAYERED FILM SUBJECTED TO ULTRAFAST LASER HEATING

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In the paper, the mathematical model, numerical algorithm and examples of computations concerning thermal processes proceeding in a multi-layered thin film subjected to an ultrafast laser pulse are discussed. The equations describing a course of the analysed process correspond to the dual-phase-lag model and contain both the relaxation time τ_q and additionally the thermalization time τ_T . At the stage of numerical simulation, the finite difference method has been used. The algorithm is based on an artificial decomposition of the domain considered, while common thermal interactions between successive layers are taken into account using conditions of heat flux and temperature continuity at points corresponding to internal boundaries (1D task has been considered).

Key words: microscale heat transfer, thin films, laser pulse, numerical modelling

1. Introduction

Classical Fourier's equation constitutes a quite good mathematical description of heat conduction processes proceeding in macro domains subjected to external thermal interactions whose duration is not very short, at the same time the temperature considered T(x,t) should be essentially bigger than 0 K (Al-Nimr, 1997; Chen *et al.*, 2004; Escobar *et al.*, 2006; Özişik and Tzou, 1994; Tamma and Zhou, 1998). It is well known that the Fourier law assumes instantaneous heat propagation and this assumption leads to evident errors when the time considered is comparable with the relaxation time τ_q of heat carriers (average time between successive electron-phonon collisions in the conductors or semiconductors and phonon-phonon collisions in dielectrics). Additionally, the Fourier equation is acceptable when the characteristic dimension L of the domain considered is essentially larger than the mean free path Λ of the heat carriers (the average distance that energy carriers travel between successive collisions). So, generally speaking, considering the processes proceeding in domains for which $L \leq \Lambda$ (e.g. thin films) subjected to ultrafast heating (e.g. short-pulse laser interaction) other models of heat transport phenomena must be taken into account. The limitations concerning the Fourier model applications are discussed, among others, in Escobar *et al.* (2006), Özişik and Tzou (1994), Tamma and Zhou (1998).

In the paper, the problem of heat transfer proceeding in a multi-layered thin film subjected to a short pulse laser heating is considered. It should be pointed out that the heat transport through thin films is of vital importance in microtechnology applications (Dai and Nassar, 2001a,b; Smith and Norris, 2003). The mathematical description of the process discussed is based on the dual-phase-lag model presented, among others, in Escobar *et al.* (2006), Özişik and Tzou (1994), Tamma and Zhou (1998), Tzou (1995). Taking into account characteristic features of thin film geometry one can assume that the components of heat flux in macro-directions (e.g. x_2, x_3) result from the traditional Fourier law, while in the definition of heat flux in the direction x_1 the relaxation time τ_q and additionally the thermalization time τ_T (the mean time required for electrons and lattice to reach equilibrium) are introduced (Dai and Nassar, 2001). So, in this direction the heat flux and temperature gradient will occur at different times.

The mathematical model presented in the next section concerns a 1D problem corresponding to the micro-direction $x_1 = x$ (heat fluxes in the directions x_2 , x_3 are neglected). For most short laser pulse interactions with thin films, the laser spot size is much larger than the film thickness. Therefore, it is reasonable to treat the interactions as a one-dimensional heat transfer process (Chen and Beraun, 2001).

At the stage of numerical realisation, an algorithm based on the finite difference method is applied, at the same time a certain concept of domain decomposition is proposed. Temporary temperature fields in successive layers are calculated separately, while the continuity conditions allow one to find the temporary solution concerning the entire domain. In the final part of the paper, examples of computations are shown.

2. Heat transport at the microscale

Heat transport equations describing thermal behaviour of a thin film, as shown in Fig. 1, can be written in the form (Dai and Nassar, 2000; Dai and Nassar, 2001; Özişik and Tzou, 1994; Tamma and Zhou, 1998; Tzou, 1995)

$$c\frac{\partial T(x,t)}{\partial t} = -\nabla \cdot \boldsymbol{q}(x,t) + Q(x,t)$$
(2.1)

and

$$q_1(x, t + \tau_q) = -\lambda \frac{\partial T(x, t + \tau_T)}{\partial x_1}$$

$$q_2(x, t) = -\lambda \frac{\partial T(x, t)}{\partial x_2} \qquad q_3(x, t) = -\lambda \frac{\partial T(x, t)}{\partial x_3}$$
(2.2)

where $x = \{x_1, x_2, x_3\}$, $\boldsymbol{q} = [q_1, q_2, q_3]^{\top}$ is the heat flux, λ is the thermal conductivity, c is the volumetric specific heat, Q is the capacity of internal heat sources, τ_T , τ_q are the positive constants which are the time lags of the temperature gradient and heat flux, respectively.



Fig. 1. Thin film

Using the Taylor series expansion, the following first-order approximation of equation $(2.2)_1$ can be taken into account

$$q_1(x,t) + \tau_q \frac{\partial q_1(x,t)}{\partial t} = -\lambda \Big[\frac{\partial T(x,t)}{\partial x_1} + \tau_T \frac{\partial}{\partial t} \Big(\frac{\partial T(x,t)}{\partial x_1} \Big) \Big]$$
(2.3)

Equation (2.1), which in the case of 1D problem $(x = x_1)$ is of the form

$$c\frac{\partial T(x,t)}{\partial t} = -\frac{\partial q(x,t)}{\partial x} + Q(x,t)$$
(2.4)

where

$$q(x,t) + \tau_q \frac{\partial q(x,t)}{\partial t} = -\lambda \Big[\frac{\partial T(x,t)}{\partial x} + \tau_T \frac{\partial}{\partial t} \Big(\frac{\partial T(x,t)}{\partial x} \Big) \Big]$$
(2.5)

should be supplemented by adequate boundary and initial conditions.

3. Multi-layered domain

Let us consider a multi-layered thin film of thickness $L = L_1 + L_2 + \ldots + L_M$ (as in Fig. 2) with the initial temperature distribution $T(x, 0) = T_0$, constant thermal properties of successive layers, ideal thermal contact between the layers and insulated external boundaries. The front surface x = 0 is irradiated by a laser pulse whose output intensity equals I(t). According to Tang and Araki (1999), the conductional heat transfer can be modeled by equation (2.4) with internal volumetric heat sources Q(x,t), at the same time for x = 0 the non-flux condition can be assumed. In this paper, the following formula (Kaba and Dai, 2005; Tzou and Chiu, 2001) has been applied

$$Q(x,t) = \sqrt{\frac{\beta}{\pi} \frac{1-R}{t_p \delta}} I_0 \exp\left(-\frac{x}{\delta} - \sqrt{\beta} \frac{|t-2t_p|}{t_p}\right)$$
(3.1)

where I_0 is the laser intensity which is defined as the total energy carried by the laser pulse per unit cross-section of the laser beam, t_p is the characteristic time of the laser pulse, δ is the characteristic transparent length of irradiated photons called the absorption depth, R is the reflectivity of the irradiated surface and $\beta = 4 \ln 2$ (Chen and Beraun, 2001).

The local and temporary value of Q(x,t) results from the distance x between the surface subjected to laser action and the point considered. So, the following system of equations is taken into account

$$x \in \Omega_m: \qquad c_m \frac{\partial T_m(x,t)}{\partial t} = -\frac{\partial q_m(x,t)}{\partial x} + Q_m(x,t) \qquad m = 1, 2, \dots, M$$

$$(3.2)$$

$$q_m(x,t) + \tau_{qm} \frac{\partial q_m(x,t)}{\partial t} = -\lambda_m \Big[\frac{\partial T_m(x,t)}{\partial x} + \tau_{Tm} \frac{\partial}{\partial t} \Big(\frac{\partial T_m(x,t)}{\partial x} \Big) \Big]$$

The boundary conditions on the contact surfaces between the sub-domains have the form of continuity ones, which means

$$x \in \Gamma_m: \begin{cases} T_m(x,t) = T_{m+1}(x,t) \\ q_m(x,t) = q_{m+1}(x,t) \end{cases} \qquad m = 1, 2, \dots, M-1 \qquad (3.3)$$



Fig. 2. Multi-layered domain

The initial conditions are assumed in the following way

$$t = 0:$$
 $T_m(x, 0) = T_{m0}$ $\frac{\partial T_m(x, t)}{\partial t}\Big|_{t=0} = 0$ (3.4)

4. Numerical model

At the stage of numerical computations, the finite difference method has been used, while the final system of equations has been solved using the Thomas algorithm (Majchrzak and Mochnacki, 2004; Mochnacki and Suchy, 1995) (separately for successive layers).

To find a numerical solution to the problem discussed, a staggered grid is introduced (Dai and Nassar, 2000), as shown in Fig. 3. For convenience, we omit m and denote $T_i^f = T(ih, f\Delta t)$, where h is the mesh size, Δt is the time step, $i = 0, 2, 4, \ldots, N$, $f = 0, 1, \ldots, F$, and $q_j^f = q(jh, f\Delta t)$, where $j = 1, 3, \ldots, N - 1$.



Fig. 3. Discretization

As was mentioned, the numerical procedure proposed is based on the Thomas algorithm for a tridiagonal linear system of equations and decomposition of the domain into M sub-domains corresponding to successive layers. Additionally, an adequate procedure of contact temperatures computations is introduced.

Let us consider an internal point $x_i \in \Omega_m$. The finite difference approximation of equation $(3.2)_1$ can be written as follows (implicit scheme)

$$c_i \frac{T_i^f - T_i^{f-1}}{\Delta t} = -\frac{q_{i+1}^f - q_{i-1}^f}{2h} + Q_i$$
(4.1)

where the index *i* corresponds to 'temperature nodes' (Fig. 3) belonging to the Ω_m sub-domain.

Equation $(3.2)_2$ can be transformed to the form

$$q_{j}^{f} + \tau_{qj} \frac{q_{j}^{f} - q_{j}^{f-1}}{\Delta t} = -\lambda_{j} \left(\frac{T_{j+1}^{f} - T_{j-1}^{f}}{2h} \right) - \frac{\lambda_{j} \tau_{Tj}}{\Delta t} \left(\frac{T_{j+1}^{f} - T_{j-1}^{f}}{2h} - \frac{T_{j+1}^{f-1} - T_{j-1}^{f-1}}{2h} \right)$$
(4.2)

or

$$q_{j}^{f} = \frac{\tau_{qj}}{\Delta t \left(1 + \frac{\tau_{qj}}{\Delta t}\right)} q_{j}^{f-1} - \frac{\lambda_{j} \left(1 + \frac{\tau_{Tj}}{\Delta t}\right)}{2h \left(1 + \frac{\tau_{qj}}{\Delta t}\right)} (T_{j+1}^{f} - T_{j-1}^{f}) + \frac{\lambda_{j} \tau_{Tj}}{2h \Delta t \left(1 + \frac{\tau_{qj}}{\Delta t}\right)} (T_{j+1}^{f-1} - T_{j-1}^{f-1})$$

$$(4.3)$$

where the index j corresponds to 'heat flux nodes' (Fig. 3) belonging to the Ω_m sub-domain.

The last equation allows one to construct similar formulas for the nodes i - 1, i + 1, and then one obtains ($\tau_{qi} = \tau_{qi-1} = \tau_{qi+1}$, $\tau_{Ti} = \tau_{Ti-1} = \tau_{Ti+1}$, $\lambda_i = \lambda_{i-1} = \lambda_{i+1}$, of course)

$$q_{i-1}^{f} - q_{i+1}^{f} = \frac{\tau_{qi}}{\Delta t \left(1 + \frac{\tau_{qi}}{\Delta t}\right)} \left(q_{i-1}^{f-1} - q_{i+1}^{f-1}\right) + \frac{\lambda_{i} \left(1 + \frac{\tau_{Ti}}{\Delta t}\right)}{2h \left(1 + \frac{\tau_{qi}}{\Delta t}\right)} \left(T_{i-2}^{f} - 2T_{i}^{f} + T_{i+2}^{f}\right) + \frac{\lambda_{i} \left(1 + \frac{\tau_{qi}}{\Delta t}\right)}{2h \left(1 + \frac{\tau_{qi}}{\Delta t}\right)} \left(T_{i-2}^{f-1} - 2T_{i}^{f-1} + T_{i+2}^{f-1}\right)$$

$$(4.4)$$

Putting (4.4) into (4.1), one has

$$c_{i}\frac{T_{i}^{f} - T_{i}^{f-1}}{\Delta t} = \frac{\tau_{qi}}{2h\Delta t \left(1 + \frac{\tau_{qi}}{\Delta t}\right)} (q_{i-1}^{f-1} - q_{i+1}^{f-1}) + \frac{\lambda_{i} \left(1 + \frac{\tau_{Ti}}{\Delta t}\right)}{4h^{2} \left(1 + \frac{\tau_{qi}}{\Delta t}\right)} (T_{i-2}^{f} - 2T_{i}^{f} + T_{i+2}^{f}) + \frac{\lambda_{i} \tau_{Ti}}{4h^{2} \Delta t \left(1 + \frac{\tau_{qi}}{\Delta t}\right)} (T_{i-2}^{f-1} - 2T_{i}^{f-1} + T_{i+2}^{f-1}) + Q_{i}^{f}$$

$$(4.5)$$

or

$$A_i T_{i-2}^f - (1+2A_i) T_i^f + A_i T_{i+2}^f = D_i^f$$
(4.6)

where

$$A_{i} = \frac{\lambda_{i} \Delta t \left(1 + \frac{\tau_{T_{i}}}{\Delta t}\right)}{4h^{2} c_{i} \left(1 + \frac{\tau_{q_{i}}}{\Delta t}\right)}$$
(4.7)

and

$$D_{i}^{f} = B_{i}T_{i-2}^{f-1} - (1+2B_{i})T_{i}^{f-1} + B_{i}T_{i+2}^{f-1} + C_{i}(q_{i+1}^{f-1} - q_{i-1}^{f-1}) - \frac{\Delta t}{c_{i}}Q_{i}^{f} \quad (4.8)$$

while

$$B_i = \frac{\lambda_i \tau_{T_i}}{4h^2 c_i \left(1 + \frac{\tau_{q_i}}{\Delta t}\right)} \qquad C_i = \frac{\tau_{q_i}}{2hc_i \left(1 + \frac{\tau_{q_i}}{\Delta t}\right)}$$
(4.9)

Let us assume that the contact temperatures $T_i^f = T_{cm}^f$ at the boundary points $x_m, m = 1, 2, ..., M - 1$ are known. Then the temperature field at the time t^f results from the following systems of equations: — first layer

$$T_0^f - T_2^f = \frac{\frac{\tau_{T1}}{\Delta t}}{1 + \frac{\tau_{T1}}{\Delta t}} (T_2^{f-1} - T_0^{f-1})$$

$$A_i T_{i-2}^f - (1 + 2A_i) T_i^f + A_i T_{i+2}^f = D_i^f \qquad i = 2, 4, \dots, N_1 - 2 \quad (4.10)$$

$$T_{N_1}^f = T_{c1}^f$$

— internal layers

$$T_{N_{m-1}}^{f} = T_{cm-1}^{f}$$

$$A_{i}T_{i-2}^{f} - (1+2A_{i})T_{i}^{f} + A_{i}T_{i+2}^{f} = D_{i}^{f}$$

$$i = N_{m-1} + 2, N_{m-1} + 4, \dots, N_{m} - 2 \qquad (4.11)$$

$$T_{N_{m}}^{f} = T_{cm}^{f}$$

— last layer

$$T_{N_{M-1}}^{f} = T_{cM-1}^{f}$$

$$A_{i}T_{i-2}^{f} - (1+2A_{i})T_{i}^{f} + A_{i}T_{i+2}^{f} = D_{i}^{f}$$

$$i = N_{M-1} + 2, N_{M-1} + 4, \dots, N-2 \qquad (4.12)$$

$$T_{N-2}^{f} - T_{N}^{f} = \frac{\frac{\tau_{TM}}{\Delta t}}{1 + \frac{\tau_{TM}}{\Delta t}} (T_{N}^{f-1} - T_{N-2}^{f-1})$$

Finally, the problem of computations of contact temperatures will be explained. The continuity condition $q_m(x,t) = q_{m+1}(x,t) = q_{cm}(x,t)$, formula (3.3), leads to the equation $x \in \Gamma_m$

$$\tau_{qm} \frac{\partial q_m(x,t)}{\partial t} + \lambda_m \frac{\partial T_m(x,t)}{\partial x} + \lambda_m \tau_{Tm} \frac{\partial^2 T_m(x,t)}{\partial t \partial x} =$$

$$= \tau_{qm+1} \frac{\partial q_{m+1}(x,t)}{\partial t} + \lambda_{m+1} \frac{\partial T_{m+1}(x,t)}{\partial x} + \lambda_{m+1} \tau_{Tm+1} \frac{\partial^2 T_{m+1}(x,t)}{\partial t \partial x}$$

$$(4.13)$$

This formula should be written down using the finite difference convention, and then

$$\alpha_m T_{cm}^f = \lambda_m \left(1 + \frac{\tau_{Tm}}{\Delta t} \right) T_{N_m-2}^f + \lambda_{m+1} \left(1 + \frac{\tau_{Tm+1}}{\Delta t} \right) T_{N_m+2}^{f-1} + \frac{\lambda_m \tau_{Tm}}{\Delta t} (T_{cm}^{f-1} - T_{N_m-2}^{f-1}) + \lambda_m \tau_{Tm} (T_{cm}^{f-1} - T_{N_m-2}^{f-1}) + (4.14) + \lambda_{m+1} \tau_{Tm+1} (T_{cm}^{f-1} - T_{N_m+2}^{f-1}) + \frac{2h}{\Delta t} (\tau_{qm+1} - \tau_{qm}) (q_{cm}^f - q_{cm}^{f-1})$$

where

$$\alpha_m = \lambda_m \left(1 + \frac{\tau_{Tm}}{\Delta t} \right) + \lambda_{m+1} \left(1 + \frac{\tau_{Tm+1}}{\Delta t} \right)$$
(4.15)

In the place of q_{cm}^f and q_{cm}^{f-1} , the arithmetic means of heat flux values at the points N_{m-1} , N_{m+1} are introduced. The starting point of computations consists in assumption that $T_{cm}^0 = T_{cm}^1 = T_0$ and $q_{cm}^0 = 0$. Next, the system of equations (4.10), (4.11), (4.12) is solved and the heat fluxes at the odd internal nodes are found by means of equation (4.3). Finally, the contact temperatures T_{cm}^f are calculated using formula (4.14) and the next loop of computations can be realised. The method proposed is very quick and effective owing to application of the Thomas algorithm and decomposition of the domain.

5. Results of computations

To test the accuracy and effectiveness of the method proposed, at first the following task has been solved. The layer of thickness $L = 10^{-4}$ and thermophysical parameters $\lambda = 1$, c = 1, $\tau_q = 1/\pi^2 + 100$, $\tau_T = 1/\pi^2 + 10^{-6}$, Q(x,t) = 0 has been considered. For the data assumed, the problem described by equations (2.4), (2.5) and boundary-initial conditions in the form

$$T(0,t) = 0 T(L,t) = 0 (5.1)$$
$$T(x,0) = \sin(10^4 \pi x) \frac{\partial T(x,t)}{\partial t}\Big|_{t=0} = -\pi^2 \sin(10^4 \pi x)$$

has the following analytical solution (Dai and Nassar, 2001)

$$T(x,t) = \exp(-\pi^2 t) \sin(10^4 \pi x)$$
(5.2)

So, the domain considered has been divided in an artificial way into 4 parts of the same thickness, and ideal thermal contact between the sub-domains has been assumed. Using the algorithm presented in the previous sections on the assumption that $h = 5 \cdot 10^{-7}$ and $\Delta t = 0.0001$, the transient temperature field has been found and the results have been compared with the exact solution. Both solutions are very close as shown in Fig. 4.



Fig. 4. Analytical (lines) and numerical (symbols) solutions

The second task is connected with the numerical solution presented in Dai and Nassar (2000) which concerns a two-layer domain (gold layer and chromium layer of thicknesses 50 nm). In order to test the algorithm discussed, the domain considered has been divided into 4 parts (Ω_1 and Ω_2 correspond to the gold sub-domain, Ω_3 and Ω_4 correspond to the chromium sub-domain). The layers are subjected to short-pulse laser irradiation (R = 0.93, $I_0 = 13.7 \text{ J/m}^2$, $t_p = 100 \text{ fs}$, $\delta = 15.3 \text{ nm}$). Thermophysical parameters of the sub-domains are the following: $\lambda = 317 \text{ W/(mK)}$, $c = 2.4897 \text{ MJ/(m^3K)}$, $\tau_q = 8.5 \text{ ps}$ ($1 \text{ ps}=10^{-12} \text{ s}$), $\tau_T = 90 \text{ ps}$ (gold), $\lambda = 93 \text{ W/(mK)}$, $c = 3.2148 \text{ MJ/(m^3K)}$, $\tau_q = 0.136 \text{ ps}$, $\tau_T = 7.86 \text{ ps}$ (chromium). The mesh step: h = 1 nm, time step: $\Delta t = 0.005 \text{ ps}$.

In Fig. 5, the temperature profiles (temperature rise above $T_0 = 27^{\circ}$ C) for the instants 0.2 ps and 0.25 ps are shown. The results of both solutions are close. The temperatures obtained using the algorithm presented here are bigger, indeed. It results from the fact that the laser interaction was probably approximated in a little different way, additionally the approach to the continuity conditions and the concept of decomposition were different, too.



Fig. 5. Temperature profiles – comparison with solution (symbols) presented in Dai and Nassar (2000)

The last example concerns the alternating gold-chromium-gold-chromium layers. Thermophysical parameters of the materials are the same as previously, the laser characteristic is also the same.

In Fig. 6 the temperature profiles (temperature rise above $T_0 = 27^{\circ}$ C) for 1 - 0.4 ps, 2 - 0.6 ps, 3 - 0.8 ps and 4 - 1 ps are shown. Figure 7 illustrates the course of temperature at the surface subjected to laser heating (x = 0) and the internal surfaces $x = L_1$, $x = L_1 + L_2$.



Fig. 6. Temperature profiles in the multi-layer domain



Fig. 7. Heating (cooling) curves at points selected from the domain Ω

6. Final remarks

The presented model based on the dual-phase-lag approach contains both the relaxation time τ_q and additionally the thermalization time τ_T . In literature concerning the microscale heat transfer, one can also find models for which only the relaxation time is taken into account. In this place the well known Cataneo equation can be mentioned. According to present opinions resulting mainly from experiments (Özişik and Tzou, 1994; Tank and Araki, 1999), it

seems that the assumption concerning a non-zero value of τ_T gives results closer to real physical conditions of the microscale heat transfer.

The algorithm presented can be simply generalised for 2D or 3D tasks. The components determining $q_2(x,t)$ and $q_3(x,t)$ result then directly from the classical Fourier law. A numerical solution obtained in this way gives a possibility to analyse the influence of laser pulse distribution in the directions x_2 and x_3 on the course of heating and cooling processes in the domain considered.

The model presented here can be used for analysis of a heat transfer proceeding in multi-layered domains being a composition of an optional number of thin films with different parameters. The choice of materials considered in this paper results, first of all, from the available in literature input data. It seems that more close to real thermal conditions is the 2D model corresponding to an axially symmetrical domain, and this problem will be a subject of the future research.

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Symulacja numeryczna procesów cieplnych zachodzących w wielowarstwowych mikroobszarach poddanych działaniu ultrakrótkiego impulsu laserowego

Streszczenie

W pracy przedstawiono model matematyczny, algorytm numeryczny i przykłady symulacji dotyczących przebiegu procesów cieplnych w wielowarstwowym mikroobszarze nagrzewanym ultraszybkim impulsem generowanym przez laser. Równanie opisujące przebieg procesu odpowiada modelowi z dwoma opóźnieniami wynikającymi z czasu relaksacji i czasu termalizacji. Na etapie obliczeń numerycznych wykorzystano metodę różnic skończonych. Algorytm bazuje na sztucznej dekompozycji obszaru wielowarstwowego, przy czym wzajemne oddziaływania między warstwami uwzględniono poprzez założenie ciągłości strumienia ciepła i pola temperatury na powierzchniach kontaktu. Biorąc pod uwagę geometrię obszaru, rozpatrywano zadanie jednowymiarowe.

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