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Identification of electron-phonon coupling factor in a thin metal film subjected to an ultrashort laser pulse

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A thin metal film subjected to a laser pulse is considered. The problem is described by the system of energy equations describing the electron gas and lattice temperatures. The thermal interactions between electrons and lattice are determined by the parameter G called the electron-phonon coupling factor. To estimate the unknown parameter G the identification problem is formulated. The additional information necessary to solve an inverse problem is the knowledge of transient measurements of the reflectivity or transmissivity variation which is proportional to the variation of the electron temperature. So, at the stage of inverse problem solution, it is possible to assume the knowledge of electrons temperature on the irradiated surface of the system ($x = 0$). To solve the identification problem the gradient method basing on the least squares criterion and sensitivity coefficients is used. In the final part of the paper the results of computations are shown.

Keywords: microscale heat transfer, laser heating, two-temperature model, inverse problem, finite difference method.

1. Introduction

Analysis of thermal problems connected with the materials processing technology design requires, among others, to find the solutions of heat conduction problems in a microscale when the Fourier law cannot be applied. The differences between the macroscopic energy equation basing on the Fourier law and the models describing the microscale heat transfer appear because of the extremely short duration, the extreme temperature gradients and the very small geometrical dimensions of an object considered. This situation takes place, among others, during a short-pulse laser heating of thin metal films and the different models are used to describe the non-Fourier heat conduction [2, 5, 8, 10, 16]. One of them is the hyperbolic two-temperature model [1, 11, 15, 16] determining the temporal and spatial evolution of the lattice and electrons temperatures and this approach is here used. The thermal interactions between electrons temperature T_e and phonons temperature T_l are determined by the parameter G called a coupling factor [8]. The assumption that the values of T_e and T_l are not big allows one to treat the parameter G as a constant value and a such situation is considered below. The laser heating is taken into account by the introduction of internal heat source appearing in the equation determining the course of electrons temperature.

The additional information necessary to solve the inverse problem is the knowledge of transient measurements of the variation of reflectivity which is proportional to the variation of the electrons temperature [1]. So, at the stage of inverse problem formulation , the knowledge of electrons temperature on the irradiated surface of the system ($x = 0$) can be assumed. To solve the identification problem the gradient method basing on the least squares criterion and sensitivity coefficients has been applied [7, 13]. The basic problem and additional one connected with the sensitivity analysis are solved using the explicit scheme of finite difference method. In the final part of the paper the results of identification are shown both in the case when the real measurements quoted in literature have been used and also when the direct problem solution has been applied.

2. Direct problem

The 1D problem is analyzed, this means that the heat transfer in the direction perpendicular to the external surfaces of metal film is considered, while the front surface $x = 0$ is irradiated by a laser pulse. Taking into account the geometry of the domain considered (Fig. 1) this assumption is quite acceptable.

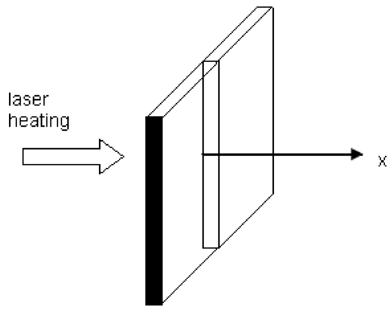

Fig. 1. Thin film.

Hyperbolic two-temperature model describing the temporal and spatial evolution of the lattice and electrons temperatures (T_l and T_e) in the irradiated metal can be written in a form of two coupled nonlinear differential equations [1, 11]

$$C_e(T_e)\frac{\partial T_e(x,t)}{\partial t} = -\frac{\partial q_e(x,t)}{\partial x} - G(T_e)\left[T_e(x,t) - T_l(x,t)\right] + Q(x,t), \tag{1}$$

$$C_l(T_l)\frac{\partial T_l(x,t)}{\partial t} = -\frac{\partial q_l(x,t)}{\partial x} + G(T_e)\left[T_e(x,t) - T_l(x,t)\right], \tag{2}$$

where $C_e(T_e)$, $C_l(T_l)$ are the volumetric specific heats of the electrons and lattice, respectively, G is the electron-phonon coupling factor related to the rate of energy exchange between electrons and lattice, $q_e(x,t)$, $q_l(x,t)$ are the heat fluxes and $Q(x,t)$ is the source term connected with the laser action.

In a place of classical Fourier law the following formulas are introduced

$$q_e(x,t+\tau_e) = -\lambda_e(T_e,T_l)\frac{\partial T_e(x,t)}{\partial x}, \tag{3}$$

$$q_l(x,t+\tau_l) = -\lambda_l(T_l)\frac{\partial T_l(x,t)}{\partial x}, \tag{4}$$

where $\lambda_e(T_e,\ T_l)$, $\lambda_l(T_l)$ are the thermal conductivities of the electrons and lattice, respectively, τ_e is the relaxation time of free electrons in metals (the mean time for electrons to change their states), τ_l is the relaxation time in phonon collisions.

The internal heat source $Q(x,t)$ connected with the laser action is given in the form [1]

$$Q(x,t) = \sqrt{\frac{\beta}{\pi}}\frac{1-R}{t_p\delta}I_0\exp\left[-\frac{x}{\delta} - \beta\frac{(t-2t_p)^2}{t_p^2}\right], \tag{5}$$

where I_0 is the laser intensity, t_p is the characteristic time of laser pulse, δ is the optical penetration depth, R is the reflectivity of the irradiated surface and $\beta = 4\ln 2$ [2].

Taking into account the short period of laser heating, the heat losses from the front and back surfaces of thin film can be neglected [1], this means

$$q_e(0,t) = q_e(L,t) = q_l(0,t) = q_l(L,t) = 0, \tag{6}$$

where L is the film thickness.

The initial conditions are assumed to be the constant ones

$$t = 0\colon T_e(x,0) = T_l(x,0) = T_p. \tag{7}$$

To define the thermal conductivity λ_e and heat capacity C_e of electrons the following formulas are widely used [1, 8, 11, 15]

$$\lambda_e(T_e, T_l) = \lambda_0 \frac{T_e}{T_l}, \tag{8}$$

$$C_e(T_e) = \gamma T_e, \tag{9}$$

where λ_0, γ are the material constants.

It should be pointed out that the simple form of dependences (8), (9) is only suitable for temperatures T_e much smaller than the Fermi temperature $T_F = E_F/k_B$, at the same time E_F, k_B are the Fermi energy and Boltzmann constant, respectively [8].

Taking into account the dependences (8), (9) and assuming the constant values of the others thermophysical parameters the Eqs. (1) and (2) can be written in the form

$$C_e(T_e)\frac{\partial T_e(x,t)}{\partial t} = -\frac{\partial q_e(x,t)}{\partial x} - G\left[T_e(x,t) - T_l(x,t)\right] + Q(x,t), \tag{10}$$

$$C_l\frac{\partial T_l(x,t)}{\partial t} = -\frac{\partial q_l(x,t)}{\partial x} + G\left[T_e(x,t) - T_l(x,t)\right]. \tag{11}$$

Using the Taylor series expansion the following first-order approximation of Eqs. (3), (4) is introduced

$$q_e(x,t) + \tau_e\frac{\partial q_e(x,t)}{\partial t} = -\lambda_e(T_e, T_l)\frac{\partial T_e(x,t)}{\partial x}, \tag{12}$$

$$q_l(x,t) + \tau_l\frac{\partial q_l(x,t)}{\partial t} = -\lambda_l\,\frac{\partial T_l(x,t)}{\partial x}. \tag{13}$$

Solving the direct problem one assumes that the thermophysical parameters appearing in Eqs. (10)–(13) are known and the electrons and lattice temperature can be found on the basis of boundary-initial problem above formulated.

3. Inverse problem

If the parameters appearing in governing equations are known then the direct problem is analyzed, while if part of them is unknown then the inverse problem should be considered [7, 9, 13, 14]. In the case of microscale heat transfer the additional information necessary to solve the inverse problem corresponds to the knowledge of transient measurements of the variation of reflectivity which is proportional to the variation of the electron temperature [1]. So, at the stage of identification problem solution one can assume the knowledge of electrons temperature $T_e\,(0,t)$ at the irradiated surface of the domain, this means

$$T_{ed}^{f} = T_{ed}(0, t^f), \qquad f = 1, 2, \ldots, F. \tag{14}$$

In this paper the problem concerning the identification of electron-phonon coupling factor using gradient method [7, 13] is discussed. To solve the task, the least squares criterion is applied

$$S(G)=\frac{1}{F}\sum_{f=1}^{F}\left[T_{e0}^{f}(G)-T_{ed}^{f}\right]^{2}, \tag{15}$$

where $T_{e0}^{f}(G)=T_e(G,0,t^f)$ are the calculated electrons temperatures at the irradiated surface. This temperature is obtained from the solution of the direct problem (cf., chapter 2) with an estimate for the unknown parameter G.

The criterion (15) is differentiated with respect to the parameter considered and next the necessary condition of optimum is used

$$\frac{\mathrm{d}S}{\mathrm{d}G}=\frac{2}{F}\sum_{f=1}^{F}\left[T_{e0}^{f}(G)-T_{ed}^{f}\right]\left(U_{e0}^{f}\right)^{k}=0, \tag{16}$$

where

$$\left(U_{e0}^{f}\right)^{k}=\left.\frac{\mathrm{d}T_{e0}^{f}(G)}{\mathrm{d}G}\right|_{G=G^k} \tag{17}$$

are the sensitivity coefficients, k is the number of iteration, G^0 is the arbitrary assumed value of coupling factor, while G^k for $k>0$ results from the previous iteration.

Function $T_{e0}^{f}(G)$ is expanded in the Taylor series about known value of G^k taking into account the first derivative, this means

$$T_{e0}^{f}(G)=\left(T_{e0}^{f}(G)\right)^{k}+\left(U_{e0}^{f}\right)^{k}(G^{k+1}-G^{k}). \tag{18}$$

Introducing (18) into (16) one obtains

$$\sum_{f=1}^{F}\left(U_{e0}^{f}\right)^{k}\left(G^{k+1}-G^{k}\right)\left(U_{e0}^{f}\right)^{k}=\sum_{f=1}^{F}\left[T_{ed}^{f}-\left(T_{e0}^{f}(G)\right)^{k}\right]\left(U_{e0}^{f}\right)^{k} \tag{19}$$

or

$$G^{k+1}=G^{k}+\frac{\sum\limits_{f=1}^{F}\left[T_{ed}^{f}-\left(T_{e0}^{f}(G)\right)^{k}\right]\left(U_{e0}^{f}\right)^{k}}{\sum\limits_{f=1}^{F}\left[\left(U_{e0}^{f}\right)^{k}\right]^{2}},\quad k=0,1,2,\ldots,K, \tag{20}$$

where K is the assumed number of iterations.

4. Sensitivity analysis

To determine the sensitivity coefficients (17) the direct approach of sensitivity analysis [4, 6, 7] can be applied. So, the Eqs. (10)–(13) are differentiated with respect to the coupling factor G and then one obtains the following sensitivity model

$$\frac{\mathrm{d}C_e(T_e)}{\mathrm{d}T_e}U_e\frac{\partial T_e}{\partial t}+C_e(T_e)\frac{\partial U_e}{\partial t}=-\frac{\partial w_e}{\partial x}-(T_e-T_l)-G(U_e-U_l), \tag{21}$$

$$C_l\frac{\partial U_l}{\partial t}=-\frac{\partial w_l}{\partial x}+(T_e-T_l)+G(U_e-U_l), \tag{22}$$

$$w_e + \tau_e \frac{\partial w_e}{\partial t} = -\left[\frac{\partial \lambda_e(T_e, T_l)}{\partial T_e} U_e + \frac{\partial \lambda_e(T_e, T_l)}{\partial T_l} U_l\right] \frac{\partial T_e}{\partial x} - \lambda_e(T_e, T_l) \frac{\partial U_e}{\partial x}, \tag{23}$$

$$w_l + \tau_l \frac{\partial w_l}{\partial t} = -\lambda_l \frac{\partial U_l}{\partial x}, \tag{24}$$

where

$$\begin{aligned} U_e &= \frac{\partial T_e}{\partial G}, \qquad U_l = \frac{\partial T_l}{\partial G}, \\ w_e &= \frac{\partial q_e}{\partial G}, \qquad w_l = \frac{\partial q_l}{\partial G}. \end{aligned} \tag{25}$$

Additionally, the differentiation of Eqs. (6), (7) gives

$$w_e(0, t) = w_e(L, t) = w_l(0, t) = w_l(L, t) = 0 \tag{26}$$

and

$$t = 0\colon U_e(x, 0) = U_l(x, 0) = 0. \tag{27}$$

Summing up, the Eqs. (21)–(24) supplemented by the boundary conditions (26) and initial ones (7) create the additional problem connected with the sensitivity analysis of temperature fields T_e and T_l with respect to the coupling factor G.

The solution of the problem formulated allows one to determine, among others, the sensitivity coefficients $U_{e0} = U_e(0, t)$ at the irradiated surface $x = 0$. This information is necessary in order to solve the task using the gradient method (cf., Eq. (20)).

5. Finite difference method

To solve the basic and additional problems formulated the finite difference method is adapted. A staggered grid [3, 11] is introduced (Fig. 2). Let us denote $T_{ei}^f = T_e\ (ih,\ f\Delta t)$, $T_{li}^f = T_l\ (ih,\ f\Delta t)$, $U_{ei}^f = U_e\ (ih,\ f\Delta t)$, $U_{li}^f = U_l\ (ih,\ f\Delta t)$, where h is a mesh size, Δt is a time step, $i = 0, 2, 4, \ldots, N$, $f = 0, 1, 2, \ldots, F$, and $q_{ej}^f = q_e\ (jh,\ f\Delta t)$, $q_{lj}^f = q_l\ (jh,\ f\Delta t)$, $w_{ej}^f = w_e\ (jh,\ f\Delta t)$, $w_{lj}^f = w_l\ (jh,\ f\Delta t)$, where $j = 1, 3, \ldots, N-1$.

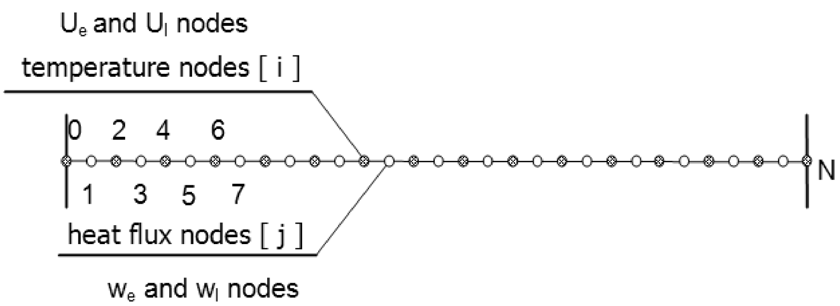

Fig. 2. Discretization.

The finite difference approximation of Eqs. (12), (13) using the explicit scheme can be written in the form

$$q_{ej}^{f-1} + \tau_e \frac{q_{ej}^f - q_{ej}^{f-1}}{\Delta t} = -\lambda_{ej}^{f-1} \frac{T_{ej+1}^{f-1} - T_{ej-1}^{f-1}}{2h} \tag{28}$$

and

$$q_{lj}^{f-1} + \tau_l \frac{q_{lj}^f - q_{lj}^{f-1}}{\Delta t} = -\lambda_l \frac{T_{lj+1}^{f-1} - T_{lj-1}^{f-1}}{2h}, \tag{29}$$

where index j corresponds to the 'heat flux nodes' (Fig. 1).

Equations (28), (29) can be transformed as follows

$$q_{ej}^f = \frac{\tau_e - \Delta t}{\tau_e} q_{ej}^{f-1} - \frac{\lambda_{ej}^{f-1} \Delta t}{2h\tau_e} \left(T_{ej+1}^{f-1} - T_{ej-1}^{f-1} \right) \tag{30}$$

and

$$q_{lj}^f = \frac{\tau_l - \Delta t}{\tau_l} q_{lj}^{f-1} - \frac{\lambda_l \Delta t}{2h\tau_l} \left(T_{lj+1}^{f-1} - T_{lj-1}^{f-1} \right). \tag{31}$$

Now, the equations (10), (11) are discretized using the explicit scheme of finite difference method

$$C_{ei}^{f-1} \frac{T_{ei}^f - T_{ei}^{f-1}}{\Delta t} = -\frac{q_{ei+1}^f - q_{ei-1}^f}{2h} - G \left(T_{ei}^{f-1} - T_{li}^{f-1} \right) + Q_i^{f-1} \tag{32}$$

and

$$C_l \frac{T_{li}^f - T_{li}^{f-1}}{\Delta t} = -\frac{q_{li+1}^f - q_{li-1}^f}{2h} + G \left(T_{ei}^{f-1} - T_{li}^{f-1} \right), \tag{33}$$

where index i corresponds to the 'temperature nodes' as shown in Fig. 1.

The dependencies (30), (31) allow ones to construct the similar formulas for nodes $i-1$, $i+1$ and then one obtains

$$q_{ei-1}^f - q_{ei+1}^f = \frac{\tau_e - \Delta t}{\tau_e} \left(q_{ei-1}^{f-1} - q_{ei+1}^{f-1} \right) + \frac{\Delta t}{2h\tau_e} \left[\lambda_{ei-1}^{f-1} \left(T_{ei-2}^{f-1} - T_{ei}^{f-1} \right) + \lambda_{ei+1}^{f-1} \left(T_{ei+2}^{f-1} - T_{ei}^{f-1} \right) \right] \tag{34}$$

and

$$q_{li-1}^f - q_{li+1}^f = \frac{\tau_l - \Delta t}{\tau_l} \left(q_{li-1}^{f-1} - q_{li+1}^{f-1} \right) + \frac{\lambda_l \Delta t}{2h\tau_l} \left(T_{li-2}^{f-1} - 2T_{li}^{f-1} + T_{li+2}^{f-1} \right). \tag{35}$$

Putting (34) into (32) and (35) into (33) one has

$$C_{ei}^{f-1} \frac{T_{ei}^f - T_{ei}^{f-1}}{\Delta t} = \frac{(\tau_e - \Delta t)}{2h\tau_e} \left(q_{ei-1}^{f-1} - q_{ei+1}^{f-1} \right) + \frac{\Delta t}{4h^2\tau_e} \left[\lambda_{ei-1}^{f-1} \left(T_{ei-2}^{f-1} - T_{ei}^{f-1} \right) + \lambda_{ei+1}^{f-1} \left(T_{ei+2}^{f-1} - T_{ei}^{f-1} \right) \right] - G \left(T_{ei}^{f-1} - T_{li}^{f-1} \right) + Q_i^{f-1} \tag{36}$$

and

$$C_l \frac{T_{li}^f - T_{li}^{f-1}}{\Delta t} = \frac{(\tau_l - \Delta t)}{2h\tau_l} \left(q_{li-1}^{f-1} - q_{li+1}^{f-1} \right) + \frac{\lambda_l \Delta t}{4h^2\tau_l} \left(T_{li-2}^{f-1} - 2T_{li}^{f-1} + T_{li+2}^{f-1} \right) + G \left(T_{ei}^{f-1} - T_{li}^{f-1} \right). \tag{37}$$

From Eqs. (36), (37) results that

$$T_{ei}^{f} = \left(1 - A_{ei}^{f-1} - B_{ei}^{f-1} - \frac{G\Delta t}{C_{ei}^{f-1}}\right) T_{ei}^{f-1} + A_{ei}^{f-1} T_{ei-2}^{f-1} + B_{ei}^{f-1} T_{ei+2}^{f-1} + \frac{G\Delta t}{C_{ei}^{f-1}} T_{li}^{f-1} + \frac{\Delta t(\tau_e - \Delta t)}{2h\tau_e C_{ei}^{f-1}} \left(q_{ei-1}^{f-1} - q_{ei+1}^{f-1}\right) + \frac{Q_i^{f-1}\Delta t}{C_{ei}^{f-1}} \tag{38}$$

and

$$T_{li}^{f} = \left(1 - 2A_{li}^{f-1} - \frac{G\Delta t}{C_l}\right) T_{li}^{f-1} + A_{li}^{f-1} \left(T_{li-2}^{f-1} + T_{li+2}^{f-1}\right) + \frac{G\Delta t}{C_l} T_{ei}^{f-1} + \frac{\Delta t(\tau_l - \Delta t)}{2h\tau_l C_l} \left(q_{li-1}^{f-1} - q_{li+1}^{f-1}\right), \tag{39}$$

where

$$A_{ei}^{f-1} = \frac{(\Delta t)^2 (\lambda_{ei-2}^{f-1} + \lambda_{ei}^{f-1})}{8h^2 \tau_e C_{ei}^{f-1}}, \qquad B_{ei}^{f-1} = \frac{(\Delta t)^2 (\lambda_{ei}^{f-1} + \lambda_{ei+2}^{f-1})}{8h^2 \tau_e C_{ei}^{f-1}}, \tag{40}$$

$$A_{li}^{f-1} = \frac{\lambda_l (\Delta t)^2}{4h^2 \tau_l C_l}. \tag{41}$$

It should be pointed out that in Eq. (30) the following approximation of thermal conductivities has been used

$$\lambda_{ej}^{f-1} = \frac{\lambda_{ej-1}^{f-1} + \lambda_{ej+1}^{f-1}}{2}. \tag{42}$$

Summing up, for transition $t^{f-1} \to t^f$ at first the Eqs. (30), (31) should be solved and next using the Eqs. (38), (39) the temperatures T_e and T_l are determined but the adequate stability criteria for explicit scheme must be fulfilled, this means (Eqs. (30), (31))

$$\frac{\tau_e - \Delta t}{\tau_e} \geq 0, \quad \frac{\tau_l - \Delta t}{\tau_l} \geq 0 \tag{43}$$

and (cf., Eqs. (38), (39))

$$1 - A_{ei}^{f-1} - B_{ei}^{f-1} - \frac{G\Delta t}{C_{ei}^{f-1}} \geq 0, \qquad 1 - 2A_{li}^{f-1} - \frac{G\Delta t}{C_l} \geq 0. \tag{44}$$

In similar way the sensitivity problem described in chapter 4 has been solved. The details of algorithm can be found in [12].

6. Results of computations

The thin gold film of thickness L ($L = 100$ nm or $L = 20$ nm) subjected to a short-pulse laser irradiation ($R = 0.93$, $I_0 = 13.4$ J/m^2, $t_p = 0.1$ ps, $\delta = 15.3$ nm – cf., Eq. (5)) [1] is considered. Thermophysical parameters are the following [8, 11]: thermal conductivities $\lambda_l = \lambda_0$, $\lambda_e = \lambda_0 T_e/T_l$, where $\lambda_0 = 315$ W/(mK), thermal capacities $C_l = 2.5$ MJ/(m^{3}K), $C_e = \gamma T_e$, where $\gamma = 70$ J/(m^{3}K^2), relaxation times $\tau_e = 0.04$ ps, $\tau_l = 0.8$ ps. Initial temperature $T_p = 300$ K.

At first, the direct problem for electron-phonon coupling factor $G = 2.6 \cdot 10^{16}$ W/(m^{3}K) has been solved using finite difference method under the assumption that $\Delta t = 0.0001$ ps and $h = 1$ nm. It should be pointed out that the calculations have been done for different time steps fulfilling the criteria of stability (cf., Eqs. (43), (44)) and the results have been practically the same.

In Figs. 3 and 4 the comparison of numerical results with experimental data presented in [1] is shown. The lines and the symbols represent calculated temperatures of electrons and experimental data, respectively. One can see that the agreement of the results obtained and measured temperatures is good.

Next, the inverse problem has been considered. In the first variant of computations the simulated measured data $T_{ed}^{f} = T_e\ (0,\ t^f)$ needed for the identification problem solution have been obtained using the values of electrons temperatures calculated from the direct problem solution under the assumption that $G = 2.6 \cdot 10^{16}$ W/(m^{3}K) (full lines in Figs. 3 and 4). Next, the inverse problem has been solved using the experimental data presented in Figs. 3 and 4 (variant 2). For all variants of computations the iteration process described by Eq. (20) has been done for $G^0 = 0$ and $K = 10$.

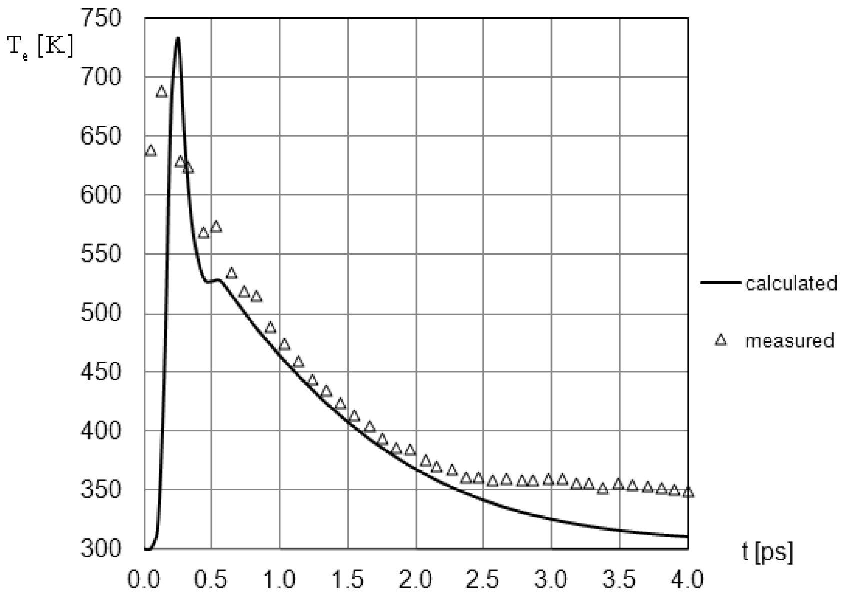

Fig. 3. Electron temperature ($x = 0$) – $L = 100$ nm.

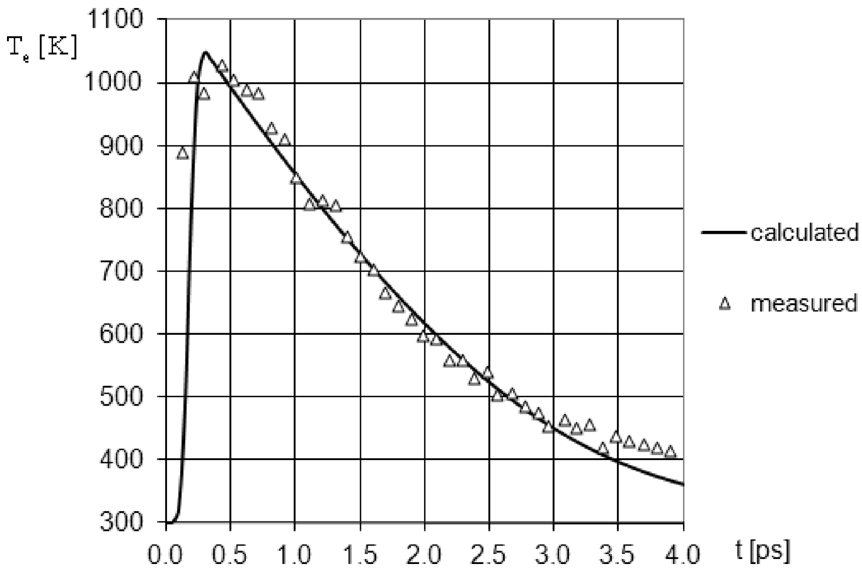

Fig. 4. Electron temperature ($x = 0$) – $L = 20$ nm.

In Figs. 5 and 6 the results of identification for 100 nm and 20 nm gold films, respectively, are shown. It is visible, that for the assumed initial value of G the iteration process is always convergent. It should be pointed out that in the case of experimental data application the estimated values of electron-phonon coupling factor G are equal to $2.668 \cdot 10^{16}$ W/(m^{3}K) for gold film of thickness $L = 100$ nm and $2.506 \cdot 10^{16}$ W/(m^{3}K) for film of thickness $L = 20$ nm, respectively. In these figures the results of identification for data disturbed by up to ± 20 K are also presented.

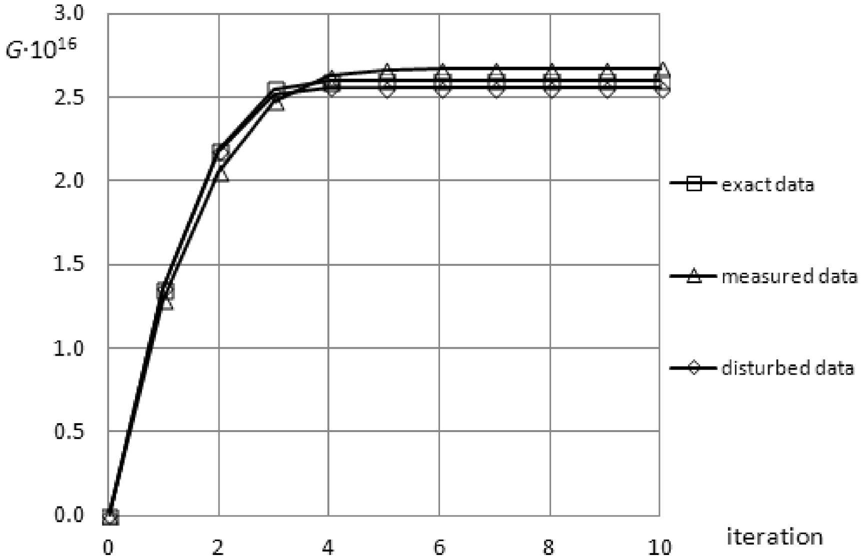

Fig. 5. Results of identification ($L = 100$ nm).

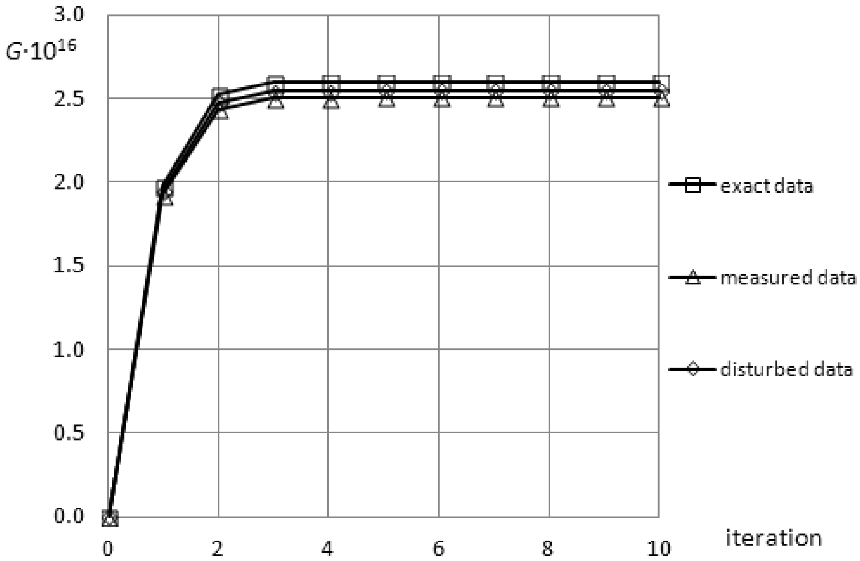

Fig. 6. Results of identification ($L = 20$ nm).

7. Final remarks

The thin metal film subjected to the ultrashort laser pulse has been considered. The thermal processes proceeding in the domain considered have been described by the hyperbolic two-temperature model in which the electron-phonon coupling factor G appears. The inverse problem connected with the identification of the parameter G has been formulated. The problem has been solved by means of the gradient method using the exact and disturbed data as well as the experimental data quoted in literature. The direct problem and the sensitivity one have been solved by means of finite difference method. It should be pointed out that the results obtained even in the case of real measurements application provide to the good estimation of coupling factor G.

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