Investigation of heat transfer in metal nanofilms irradiated with ultrashort laser pulses: two-temperature model

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Abstract. A numerical study of heat transfer between an electron gas and a crystal lattice in a metal nanofilm under irradiation with an ultrashort laser pulse was carried out on the basis of a parabolic two-temperature model of thermal conductivity presented in a dimensionless form. For the numerical solution, the finite difference method was used using the explicit-implicit Crank-Nicholson scheme. As a result of the numerical study, it was found that with an increase in the thickness of the plate, the equilibrium temperature decreases, and the time for the onset of thermal equilibrium between the electrons and the crystal lattice increases.

1. Introduction
When describing thermal conductivity in thin metal nanofilms irradiated with ultrashort laser pulses, a two-temperature model of heat transfer is used, which describes two-stage heating of the material (laser radiation excites electrons in the skin layer of a metal nanofilm, which, in turn, transfer heat to the crystal lattice) [1 – 19]. That is, this model takes into account the temperature of both the electrons and the crystal lattice. It is advisable to apply the model only for ultrashort laser pulses since in this case the electrons have time to absorb the energy from the laser radiation and are heated to high temperatures (sometimes exceeding the melting point of the material) [19], but at the same time, they do not have time to transfer heat to the crystal lattice, due to which there is a significant gradient between these two temperatures. When using longer laser pulses, the temperature gradient between the lattice and electrons is insignificant, which makes it possible to use a one-temperature model of thermal conductivity to describe the heat transfer processes.

2. The physical and mathematical formulation of the problem
In this paper, the temperature state of the electron gas and the crystal lattice of the nickel nanofilm are investigated. The nanofilm was placed on a non-thermally conductive substrate and irradiated with an ultrashort laser pulse (figure 1).
Assuming that the heat flux per unit volume is proportional to the temperature difference, we find
\[ dQ = \alpha(T_1 - T_2)dt \]
where \( T_1, T_2 \) are the temperatures of the electrons and the crystal lattice, K; \( \alpha \) is the coefficient of the volumetric heat transfer (per unit volume), W/(m\(^3\)K); \( Q \) is the heat flow per unit volume, J/m\(^3\); \( t \) is the time, s.

The heat balance equations for the electrons and crystal lattice have the form
\[
\begin{align*}
\frac{\partial T_1}{\partial t} &= -\frac{\partial q_t}{\partial x} + \alpha(T_2 - T_1) + Q(x,t) ; \\
\frac{\partial T_2}{\partial t} &= \alpha(T_1 - T_2)
\end{align*}
\]
where \( c_1, c_2 \) are the volumetric heat capacity of the electrons and crystal lattice, J/(m\(^3\)K); \( Q(x,t) \) is the volumetric heat source initiated by the laser irradiation, W/m\(^3\); \( q \) is the heat flux from electrons, W/m\(^2\); \( x \) is the coordinate, m.

The heat flux is determined by Fourier’s law equation
\[ q = -\lambda_1 \frac{\partial T_1}{\partial x} , \]
where \( \lambda_1 \) is the coefficient of thermal conductivity of electrons, W/(mK).

Substituting (4) into (2), we obtain
\[
\begin{align*}
\frac{\partial T_1}{\partial t} &= \lambda_1 \frac{\partial^2 T_1}{\partial x^2} + \alpha(T_2 - T_1) + Q(x,t) ; \\
\frac{\partial T_2}{\partial t} &= \alpha(T_1 - T_2)
\end{align*}
\]

The ration between (5) and (6) represents a system of interrelated equations for electrons and a crystal lattice. In this system, temperature equalization occurs by performing "thermal conductivity at a point", the efficiency of which is estimated by the coefficient of volumetric heat transfer. This system can be reduced to two independent equations of an electron gas and a crystal lattice [13–15]. However, in this case, relaxation coefficients appear in the mathematical formulation of the problem [13–15, 20–24], the determination of which in thermal problems is difficult, therefore, in this work, we restrict ourselves to
a simplified model. The coordinate and time dependent internal heat source arising from laser radiation is determined by the Gaussian distribution formula [7–10]

\[ Q(x,t) = Q_0 \exp \left( -\frac{x}{\mu} - \frac{(t-t_0)^2}{\beta \delta^2} \right), \quad (7) \]

where \( Q_0 = J/\mu \); \( J \) is the density of the heat flux of laser radiation, W/m²; \( \mu \) is the depth of optical penetration, m; \( t_0 \) is the time instant of the laser pulse maximum, s; \( \beta \) is the dimensionless coefficient that regulates the pulse duration.

Let us find the solution to the equations (5), (6) for an adiabatically insulated plate under the following boundary conditions

\[ T_1(x,0) = T_2(x,0) = T_0; \quad (8) \]
\[ \frac{\partial T_1(0,t)}{\partial x} = \frac{\partial T_1(\delta,t)}{\partial x} = 0, \quad (9) \]

where \( T_0 \) is the initial temperature; \( \delta \) is the plate thickness.

To reduce problems (5), (6), (8), and (9) to dimensionless form, we introduce the following dimensionless variables and parameters

\[ \Theta_1 = \frac{T_1}{T_0}; \quad \Theta_2 = \frac{T_2}{T_0}; \quad \xi = \frac{x}{\delta}; \quad \text{Fo} = \frac{\lambda_1 t}{c_1 \delta^2}; \quad \text{Fo}_0 = \frac{\lambda_1 t_0}{c_1 \delta^2}, \quad (10) \]

where \( \Theta_1, \Theta_2 \) are dimensionless temperatures; \( \xi \) is the dimensionless coordinate; \( \text{Fo} \) is the dimensionless time; \( \text{Fo}_0 \) is the dimensionless time of the laser pulse maximum.

Taking into account (10), problems (5), (6), (8), (9) become

\[ \frac{\partial \Theta_1}{\partial \text{Fo}} = \frac{\partial^2 \Theta_1}{\partial \xi^2} + F_1 \exp \left( -\frac{\xi}{\nu} - \frac{(\text{Fo} - \text{Fo}_0)^2}{\beta \text{Fo}_0^2} \right) \quad \text{Bi} (\Theta_2 - \Theta_1); \quad (11) \]
\[ \frac{\partial \Theta_2}{\partial \text{Fo}} = \text{Bi} F_2 (\Theta_1 - \Theta_2); \quad (\text{Fo} > 0; \quad 0 < \xi < 1); \quad (12) \]
\[ \Theta_1(\xi,0) = \Theta_2(\xi,0) = 1; \quad (13) \]
\[ \frac{\partial \Theta_1(0,\text{Fo})}{\partial \xi} = \frac{\partial \Theta_1(1,\text{Fo})}{\partial \xi}, \quad (14) \]

where \( F_1 = \frac{Q_0 \delta^2}{\lambda_1 T_0}; \quad F_2 = \frac{c_1}{c_2}; \quad \text{Bi} = \frac{\alpha \delta^2}{\lambda_1}; \quad \nu = \frac{\mu}{\delta}; \quad F_3 = \frac{c_1 \lambda_2}{c_2 \lambda_1}. \)

3. The numerical solution method

For the numerical solution, the finite difference method was applied using the explicit-implicit Crank Nicholson scheme (figure 2) [16]. In this case, the scheme is unconditionally stable for any ratio of \( h_{\text{Fo}} \) and \( h \).
A mixed scheme is written on the selected grid (averaging of an implicit scheme with the weight $\sigma$ and an explicit one with the weight $(1-\sigma)$). For $\sigma = 0.5$, we get the Crank-Nicholson scheme. Then equations (11) and (12) take the form

$$
\frac{\Theta_{i,j}^{t} - \Theta_{i,j}^{t-1}}{h_{\text{Fo}}} = \sigma \Theta_{i,j+1}^{t-1} - 2\Theta_{i,j}^{t-1} + \Theta_{i,j-1}^{t-1} + (1-\sigma) \left( \Theta_{i+1,j}^{t-1} - 2\Theta_{i,j}^{t-1} + \Theta_{i-1,j}^{t-1} \right) + B_i \left( \Theta_{2i,j}^{t-1} - \Theta_{i,j}^{t-1} \right) + F_i \exp \left( -\frac{\sigma h_i (i+1) + (1-\sigma) h_j}{\nu} - \frac{(\sigma h_i j - (1-\sigma) (j-1) h_j - F_{\text{Fo}})^2}{\beta F_{\text{Fo}}^2} \right),
$$

(15)

$$
\frac{\Theta_{2i,j}^{t} - \Theta_{i,j}^{t-1}}{h_{\text{Fo}}} = F_i \left( \Theta_{i,j}^{t-1} - \Theta_{2i,j}^{t-1} \right) + (1-\sigma) \left( \Theta_{i+1,j}^{t-1} - \Theta_{2i,j}^{t-1} \right). \quad (j = 1 \ldots N; \quad i = 1 \ldots M-1).
$$

(16)

Next, the terms with unknown coefficients are grouped on the time layer, and equation (15) is written in a form that is conveniently solved with the Thomas algorithm

$$
a_i \Theta_{i,j}^{t} + b_i \Theta_{i+1,j}^{t} + a_i \Theta_{i+1,j}^{t+1} = d_i,
$$

(17)

where $a_i = -\sigma h_i / h_{\text{Fo}}^2$; $b_i = h_i = M = 1 / h_{\text{Fo}} + \sigma / h_{\text{Fo}} + B_i h_i$; $M = h_{\text{Fo}} / h_{\text{Fo}} + B_i h_i$;

$$
d_i = \frac{\Theta_{i+1,j}^{t-1} - (1-\sigma) \left( \Theta_{i+1,j}^{t-1} - 2\Theta_{i,j}^{t-1} + \Theta_{i-1,j}^{t-1} \right) + B_i \left( \Theta_{2i,j}^{t-1} - \Theta_{i,j}^{t-1} \right) + F_i \exp \left( -\frac{\sigma h_i (i+1) + (1-\sigma) h_j}{\nu} - \frac{(\sigma h_i j - (1-\sigma) (j-1) h_j - F_{\text{Fo}})^2}{\beta F_{\text{Fo}}^2} \right)}{h_{\text{Fo}}}.
$$

At each time level, we obtain the system of equations (17) for $i = 1, M-1$. The system (17) is solved by the Thomas algorithm.

The system is supplemented with the initial condition (13) for $i = 0, M$

$$
\Theta_{i,0}^0 = \Theta_{2i,0}^0 = 1
$$

(18)

and then the direct course of the Thomas algorithm begins for $j = 1, N$.

For $i = 1$, the modified coefficients are as follows
\[ \gamma_i = b_i; \quad \alpha_i = \frac{-a_i}{\gamma_i}; \quad \chi_i = \frac{d_i}{\gamma_i}. \]  

(19)

For \( i = 2, M - 2 \), the modified coefficients are as follows

\[ \gamma_i = b_i + a_i \cdot \alpha_{i-1}; \quad \alpha_i = \frac{-a_i}{\gamma_i}; \quad \chi_i = \frac{d_i - a_i \chi_{i-1}}{\gamma_i}. \]  

(20)

For \( i = M - 1 \), the modified coefficients are as follows

\[ \gamma_i = b_i + a_i \cdot \alpha_{i-1}; \quad \alpha_i = 0; \quad \chi_i = \frac{d_i - a_i \chi_{i-1}}{\gamma_i}. \]  

(21)

Next, the reverse course of the Thomas algorithm begins.

For \( i = M - 1 \)

\[ \Theta_i^j = \chi_i. \]  

(22)

For \( i = M - 1, 2 \) \( \ldots \) \( i \ldots 1 \)

\[ \Theta_i^j = \alpha_i \Theta_{i+1}^j + \chi_i. \]  

(23)

After determining all of the values of \( \Theta_1 \) on the \( j \) time layer, all of the values of \( \Theta_2 \) for the \( j \) time layer are found by substituting the values found for \( \Theta_1 \) into equation (24)

\[ \Theta_2^{j+1} = \frac{\text{Bi} F_2 \left[ \sigma \Theta_2^j + (1 - \sigma) (\Theta_2^{j+1} - \Theta_2^{j+1}) \right] + \frac{\Theta_2^{j}}{h_{F_0}}}{\frac{1}{h_{F_0}} + \text{Bi} F_2 \sigma}. \]  

(24)

The values \( \Theta_{i0}^j \), \( \Theta_{iM}^j \) are found from the conditions of the adiabatic wall (14) at the boundaries \( \xi = 0 \) and \( \xi = 1 \)

\[ \Theta_{i0} = \Theta_{i1}; \]  

(25)

\[ \Theta_{iM} = \Theta_{iM-1}. \]  

(26)

Then the calculations are repeated for each \( j \) th time layer. Using the above numerical solution algorithm and the Mathcad software package, we obtain two matrices with the temperatures of \( \Theta_1 \) and \( \Theta_2 \).

4. Discussion

A numerical study was carried out for a nickel nanofilm with a thickness of \( \delta = 100 \text{ nm} \) and \( \delta = 1000 \text{ nm} \). The initial data for the calculations are presented in the table 1 [17, 18].

| Parameter | Value |
|-----------|-------|
| \( \lambda_1 \) | 90 W/(m*K) |
| \( c_1 \) | 3.2 \cdot 10^4 J/(m^3*K) |
| \( c_2 \) | 4.1 \cdot 10^6 J/(m^3*K) |
| \( J_1 \) | 60 \cdot 10^{12} W/m^2 |
| \( \mu \) | 15.3 nm |
| \( \delta \) | 100 nm |
| \( \alpha \) | 12 \cdot 10^{17} W/(m^3*K) |
| \( T_0 \) | 300 K |
| \( F_1 \) | 0.08 |
| \( F_2 \) | 13.33 |
| \( F_0 \) | 0.153 |
| \( F_0 \) | 2.8152 |
| \( \beta \) | 0.05 |
| \( t_0 \) | 10 ps |
| \( Q_0 \) | 3.9 \cdot 10^{21} J/m^3 |
The power of the laser radiation source is determined by the Gauss formula (7). A set single pulse has a maximum at the time instant \( t_0 = 10 \) ps equal to \( 3.6 \cdot 10^{21} \) W/m\(^3\), while the pulse duration is 10 ps (figure 3, 4).

![Figure 3](image1.png) ![Figure 4](image2.png)

**Figure 3.** Change in the power of laser irradiation on the surface of a nickel film.

**Figure 4.** Change in the power of laser irradiation over the thickness of the film.

The results of the performed numerical studies have shown that the energy of the laser pulse is first perceived by the electron gas, which can be heated to temperatures significantly higher than the melting point. For example, for a 100 nm thick nickel plate, the temperature on the surface \( x = 0 \) irradiated by a laser pulse was \( T_1 = 11400 \) K, which is seven times higher than the melting temperature (figure 5). Heat is transferred from the electron gas to the crystal lattice, which heats up to only 1500 K, which is due to the higher thermal inertia due to a heat capacity that is more than two orders of magnitude higher (see table 1). It should be noted that the maximum temperature in the crystal lattice is observed not at the moment of reaching the maximum of the laser pulse \( t_0 = 10 \) ps, but with a certain lag \( (t_0 = 14 \) ps for \( \delta = 100 \) nm and \( t_0 = 16 \) ps for \( \delta = 1000 \) nm).

As a result of the numerical study, it was also found that with an increase in the thickness of a nickel nanoplate from \( \delta = 100 \) nm to \( \delta = 1000 \) nm, the equilibrium temperature decreases from 885 K to 325 K (figures 5, 6). The time for the onset of thermal equilibrium between electrons and the crystal lattice increases from 245 ps for \( \delta = 100 \) nm to 3500 ps for \( \delta = 1000 \) nm.

Changes in the temperature of the electron gas and the crystal lattice over the entire thickness of the film for the case of \( \delta = 100 \) nm are shown in figures 7, 8.

![Figure 5](image3.png) ![Figure 6](image4.png)

**Figure 5.** Time-dependence of temperatures \( T_1 \) and \( T_2 \) on the surfaces of a nickel plate with a thickness of \( \delta = 100 \) nm.

**Figure 6.** Time-dependence of temperatures \( T_1 \) and \( T_2 \) on the surfaces of a nickel plate with a thickness of \( \delta = 1000 \) nm.
5. Conclusions
Using a parabolic two-temperature model of thermal conductivity, a numerical study of heat transfer between an electron gas and a crystal lattice in a metal nanofilm irradiated by an ultrashort laser pulse has been performed. It has been shown that the temperature of electrons can significantly exceed the melting point of the material, which in this case is in an unmelted state. It has been shown that with an increase in the thickness of the plate, the equilibrium temperature decreases, and the time for the onset of thermal equilibrium between the electrons and the crystal lattice increases. The time to reach the maximum temperature for a crystal lattice is longer than for an electron gas and is determined by their thermophysical properties and the coefficient of heat transfer between the electrons and the lattice.

Acknowledgments
The reported study was funded by RFBR, project number 20-38-70021.

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