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Study of Fast Relaxing Excitations Caused by Ultrashort Laser Pulses in Nanoscale Domain

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***Abstract*–By using the heat balance equation and a modified Fourier law's formula considering the relaxation of heat flow and temperature gradient, a model of the local non-equilibrium process of plate heating with ultrashort laser pulses, modelled by heat flow of time-variable second-class boundary conditions, step (discontinuous) time function, was developed. The research showed that consideration of non-locality results in the delayed plate heatup irrespective of the laser radiation flow intensity. This fact is explained by the resistance caused by the medium to the process of change of its temperature condition, which increases as the relaxation factor grows. It was also shown that in consideration of the relaxation phenomena, the boundary conditions (first-, second- and third-class) may not be fulfilled immediately - they may be set only within a definite range of the initial time section. Therefore, the immediate implementation of the boundary condition is impossible since the value of the heat transfer factor has a definite limit, depending on the relaxation properties of the medium, which may not be exceeded under any conditions of heat exchange with the medium. The relations are given which allow establishing such laser radiation flow change patterns at which the wall surface temperature would remain set and constant over time.**

Key words – Heat conduction boundary problem, local non-equilibrium heat exchange, relaxation factor, time-variable heat flow, numerical solution, analytical solution.

I. INTRODUCTION

Presently, the laser system is available allowing to generate frequency-variable light pulses with the duration of up to 10^{-15} s (fs-pulses). They may be used to create extremely non-equilibrium conditions for fast-relaxing excitations (relaxation time $10^{-11}-10^{-14}$ s), cause fast, optically induced phase transformations, implementing limiting speeds for optical processing and transfer of information, etc. Another important fact is that high power values (up to 10^{12} *W/cm*^{2}) correspond to such short duration of pulses of relatively small energy (up to 0.1 *J*), allowing one to use laser units for material processing (cutting, drilling, smelting, tempering etc.).

The known heat exchange models are based on parabolic heat conductance equations obtained without considering the relaxation factors. As a result, they include an embedded infinite heat propagation velocity which is connected with the employing the Fourier law when deriving them, where the temperature gradient (moving force being the reason) and heat flow (being the consequence) are not time-separated. Therefore, any change of the reason will immediately change the consequence. Since no infinite values of any parameters may occur in real processes, the equations derived based on the Fourier law may be adequate to real physical processes only within a particular time range. It is known that parabolic equations are inadequate in describing all the fast processes, which changing time is comparable with the relaxation time as well as temperature change at small and extrasmall time values, in any heat processes [1 – 8]. In this connection, new, more adequate mathematical models should be developed applicable to these processes. As regards the development of such models, this work elaborates the direction of considering the time-spatial non-locality, which is based on the heat flow relaxation and scalar value of the temperature gradient in the Fourier law formula. The studies of the developed model allow one to conclude that consideration of the process local non-equilibrium results in the delayed setting of boundary conditions (irrespective of their class) due to the resistance of the investigated medium to the heat penetration process. Furthermore, the higher the holding time, the higher is the value of relaxation factor. Following the results of research to maintain some set body surface temperature during heatup, the recommendations were developed for the time-change regularities of heat flows.

II. Mathematical Task Setting

For deriving the differential heat ignition equation, considering local not-equilibrium, let us represent the Fourier law formula in the form of [8]:

$$q = -\lambda \frac{\partial T}{\partial x}$$

$$q = -\lambda\left(\frac{\partial T}{\partial x} + \tau_1 \frac{\partial^2 T}{\partial x \partial t}\right) - \tau_1 \frac{\partial q}{\partial t} \qquad (1)$$

where q – heat flow; T – temperature; x – coordinate; t – time; λ – heat conductance factor; τ_1 – relaxation factor.

By substituting (1) in the heat balance equation:

$$c\rho \frac{\partial T}{\partial t} + \frac{\partial q}{\partial x} = 0 \qquad (2)$$

let us find:

$$c\rho \frac{\partial T}{\partial t} = \lambda \frac{\partial^2 T}{\partial x^2} + \lambda\tau_1 \frac{\partial^3 T}{\partial x^2 \partial t} + \tau_1 \frac{\partial}{\partial t}\left(\frac{\partial q}{\partial x}\right) \qquad (3)$$

where ρ – density; c – heat capacity.

By expressing $\frac{\partial q}{\partial x}$ of (2) and substituting in (3), one obtains:

$$\frac{\partial T}{\partial t} + \tau_1 \frac{\partial^2 T}{\partial t^2} = a\left(\frac{\partial^2 T}{\partial x^2} + \tau_1 \frac{\partial^3 T}{\partial x^2 \partial t}\right) \qquad (4)$$

where $a = \frac{\lambda}{c\rho}$ – heat conductivity factor.

It is obvious that in case of $\tau_1 = 0$, equation (4) is reduced to the classic parabolic heat conductance equation.

Let us find the solution to equation (4) under the following boundary conditions:

$$T(x, 0) = T_0 \qquad (5)$$

$$\partial T(x, 0)/\partial t = 0 \qquad (6)$$

$$-\lambda \frac{\partial T(0,t)}{\partial x} = q(t) \qquad (7)$$

$$\frac{\partial T(\delta,t)}{\partial x} = 0 \qquad (8)$$

where δ – the plate thickness; T_0 – initial temperature; $q(t) = q_0 \sin(\omega t)$ – time-variable heat flow; q_0 – heat flow oscillation amplitude; $\omega = 2\pi/\eta$ – cyclic frequency; η – full-wave oscillation period.

To represent the heat flow as a stage (discontinuous) function, let us record formula (7) as:

$$\lambda \frac{\partial T(0,t)}{\partial x} = 0 \text{ at } \sin(\omega t) < 0$$

$$-\lambda \frac{\partial T(0,t)}{\partial x} = q_0 \text{ at } \sin(\omega t) \geq 0$$

Let us introduce the following non-dimensional values and parameters:

$$\Theta = \frac{T - T_0}{T_0} \quad \mathrm{Fo} = \frac{at}{\delta^2} \quad \xi = \frac{x}{\delta} \quad \mathrm{Fo}_1 = \frac{a\tau_1}{\delta^2}$$

$$\mathrm{Ki} = \frac{q_0 \delta}{\lambda \mathrm{T}_0} \quad \mathrm{Pd} = \frac{\omega\delta^2}{a}$$

where Θ, Fo, ξ – non-dimensional temperature, time, coordinate, respectively; Ki – Kirpichev criterion; Pd – Predvoditelev criterion; Fo_1 – non-dimensional relaxation factor.

Considering the adopted designations, task (4) – (8) will be:

$$\frac{\partial\Theta(\xi,\mathrm{Fo})}{\partial \mathrm{Fo}} + \mathrm{Fo}_1 \frac{\partial^2\Theta(\xi,\mathrm{Fo})}{\partial \mathrm{Fo}^2} = \qquad (9)$$

$$= \frac{\partial^2\Theta(\xi,\mathrm{Fo})}{\partial \xi^2} + \mathrm{Fo}_1 \frac{\partial^3\Theta(\xi,\mathrm{Fo})}{\partial \xi^2 \partial \mathrm{Fo}};$$

$$(\mathrm{Fo} > 0; \quad 0 < \xi < 1)$$

$$\Theta(\xi, 0) = 1; \qquad (10)$$

$$\frac{\partial\Theta(\xi, 0)}{\partial \mathrm{Fo}} = 0; \qquad (11)$$

$$\frac{\partial\Theta(0, \mathrm{Fo})}{\partial \xi} = 0 \text{ at } \sin(\mathrm{Pd}\cdot\mathrm{Fo}) < 0 \qquad (12)$$

$$-\frac{\partial\Theta(0,\mathrm{Fo})}{\partial\xi}=\mathrm{Ki}\ \text{ at }\ \sin(\mathrm{Pd}\cdot\mathrm{Fo})\ge 0 \qquad (13)$$

$$\frac{\partial\Theta(1,\mathrm{Fo})}{\partial\xi}=0 \qquad (14)$$

III. Numerical Solving Method

To obtain solution of task (9) – (14) in the considered region, based on the finite differences method, let us introduce the temporal-spatial mesh with steps $\Delta\xi,\ \Delta\mathrm{Fo}$, respectively, by variables $\xi,\ \mathrm{Fo}$, so that:

$$\xi_k = k\Delta\xi,\quad k=\overline{1,K};\ \ \mathrm{Fo}_i = i\,\Delta\mathrm{Fo},\quad i=\overline{1,I},$$

where K, I is the number of steps by coordinates $\xi,\ \mathrm{Fo}$.

$$\frac{\Theta_k^i-\Theta_k^{i-1}}{\Delta\mathrm{Fo}}+\mathrm{Fo}_1\frac{\Theta_k^{i+1}-2\Theta_k^i+\Theta_k^{i-1}}{\Delta\mathrm{Fo}^2}=$$

$$=\frac{\Theta_{k-1}^i-2\Theta_k^i+\Theta_{k+1}^i}{\Delta\xi^2}+$$

$$+\mathrm{Fo}_1\frac{\Theta_{k-1}^i-2\Theta_k^i+\Theta_{k+1}^i-\Theta_{k-1}^{i-1}+2\Theta_k^{i-1}-\Theta_{k+1}^{i-1}}{\Delta\mathrm{Fo}\Delta\xi^2}; \qquad (15)$$

$$\Theta_k^0=0; \qquad (16)$$

$$(\Theta_k^1-\Theta_k^0)/\Delta\mathrm{Fo}=0;$$

$$\frac{\Theta_1^i-\Theta_0^i}{\Delta\xi}=0\ \text{ at }\ \sin(\mathrm{Pd}\cdot\mathrm{Fo})<0;$$

$$\frac{\Theta_1^i-\Theta_0^i}{\Delta\xi}=\mathrm{Ki}\ \text{ at }\ \sin(\mathrm{Pd}\cdot\mathrm{Fo})\ge 0; \qquad (17)$$

$$\frac{\Theta_K^i-\Theta_{K-1}^i}{\Delta\xi}=0\,. \qquad (18)$$

IV. Discussion of the Results

Fig. 1 shows the temperature calculation results for the cases when relaxation properties of materials are not considered ($\mathrm{Fo}_1=0$) and are considered ($\mathrm{Fo}_1=0.1$). Their analysis allows concluding that consideration of relaxation properties results in delayed body heatup, which causes the greatest impact at small and extrasmall values of temporal and spatial variables. As the time increases, the difference between temperature curves is reduced and with some higher values of time variable, the curves virtually coincide. This fact is the evidence that due to the thermoinertial properties of the material, the instant heatup of the body is impossible under any conditions of the external heat exchange, including from extra intensive heat flows.

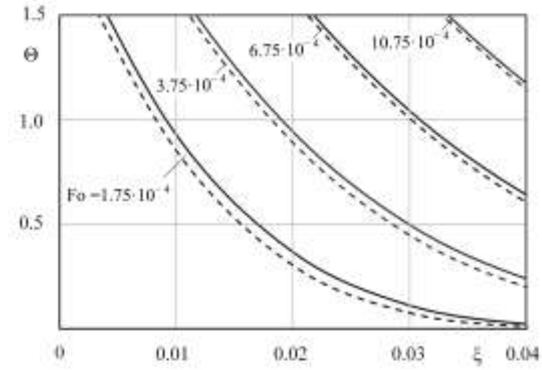

Fig. 1 Temperature distribution in plate. ———— not considering the relaxation properties of materials ($\mathrm{Fo}_1=0$); – – – – considering the relaxation properties of the material ($\mathrm{Fo}_1=0.1$); $\mathrm{Ki}=1$; $\mathrm{Pd}=1000$

Fig. 2 shows the results of calculations allowing one to evaluate the influence of heat flow discontinuity (fig. 3) on the temperature condition of the structure. It follows from their analysis that in the time range, when the heat flow is equal to zero, there is the temperature decrease close to the wall surface $(\xi=0)$, which turns to be insufficient as the coordinate rises.

Fig. 3 shows the change of heat flow over time for $\mathrm{Ki}=1.0$.

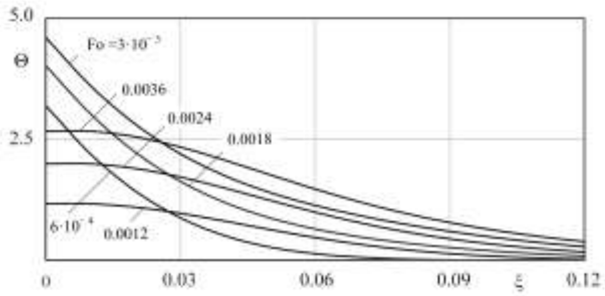

Fig. 2 Temperature distribution in plate considering the thermal flow discontinuity (fig. 3). ($\mathrm{Fo}_1=0.1$); $\mathrm{Ki}=1.0$; $\mathrm{Pd}=1000$

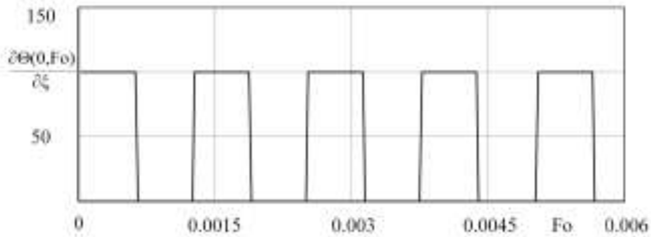

Fig. 3 Step change of heat flow over time

Fig. 4 shows the results of the calculation allowing one to estimate the value of the Kirpichev criterion by the temperature condition of the structure. It follows from their analysis that for each value of the Fo figure, there is some temperature perturbation front, which may not be exceeded by the further increase of the Kirpichev criterion (fig. 5).

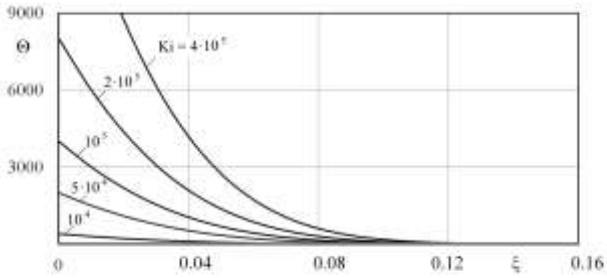

Fig. 4 Temperature distribution for $Fo_1 = 10^{-3}$ under different values of the Kirpichev criterion.

For example, for $Fo = 10^{-3}$, the value of coordinate ξ, to which the temperature perturbation front moves, increases up to $Ki = 30000$. However, a further increase of Ki causes no impact on the front value at the given value of Fo (fig.5). Consequently, the movement velocity of the heatup front movement depends on the value of the external heat souse only to a definite extent.

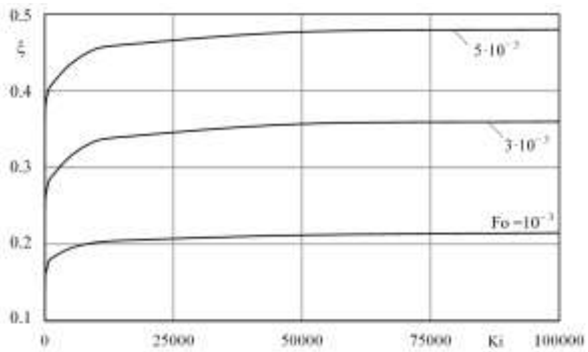

Fig. 5 Change of the temperature perturbation front value depending on the Kirpichev criterion for different values of non-dimensional time Fo

V. Method of Analytical Solution

The solutions obtained by the numerical method are inconvenient for parametrical analysis. Let us find an approximate analytical solution for task (9) - (14), by assuming $Fo_1 = 0$. The mathematical task setting in this case will be:

$$\frac{\partial\Theta(\xi, Fo)}{\partial Fo} = \frac{\partial^2\Theta(\xi, Fo)}{\partial\xi^2}; \tag{19}$$

$$(Fo > 0; \; 0 < \xi < 1)$$

$$\theta(\xi, 0) = 0; \tag{20}$$

$$-\frac{\partial\Theta(0, Fo)}{\partial\xi} = Ki(Fo); \tag{21}$$

$$\frac{\partial\Theta(1, Fo)}{\partial\xi} = 0 \tag{22}$$

The work [9] sets out the method, referring to the group of integral heat balance methods allowing obtaining analytical solutions for the non-stationary heat conductance tasks to a rather high precision practically over the whole time range of the non-stationary process $(0 < Fo < \infty)$ without any limitations for the Fourier number over the range of its small values. According to this method, the heatup process is split into two stages with reference to time: $0 < Fo \le Fo^*$ and $Fo^* \le Fo < \infty$. To this end, a time-moving boundary (temperature perturbation front) is introduced dividing the initial area $0 < \xi < 1$ into two subareas $0 \le \xi \le q_1(Fo)$ and $q_1(Fo) \le \xi \le 1$, where $q_1(Fo)$ is the function determining the boundary line movement along coordinate ξ over time (fig. 6) That is, in this case, the assumption on finite heat propagation velocity is made. In the area located beyond the temperature perturbation front, the initial temperature is preserved. The first process stages ends upon reaching coordinate $\xi = 1$, by the temperature perturbation front, i.e when $Fo = Fo^*$. At the second stage, temperature change takes place over the whole volume of the body $0 < \xi < 1$. Here additional unknown function $q_2(Fo) = \Theta(1, Fo)$ is introduced, characterizing the temperature change over time in the middle of the plate [9].

According to the above studies, consideration of the first stage of the process is of the highest interest. In this connection, the second stage will not be considered in this work. As to the first stage, due to the assumption on the finite heat propagation velocity, the heat flow moves gradually along coordinate ξ over time while having a zero value at the initial time $(q_1(0) = 0)$. Consequently, at the first process stage, the heat conductance model approaches the model assumed above (see task (4) – (8)), considering the relaxation properties of the material regardless of the fact that parabolic equation (19) is subject to solution.

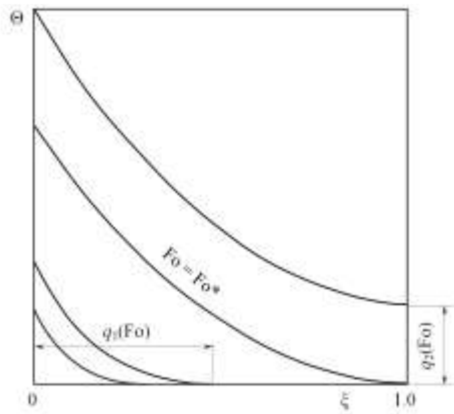

Fig. 6 Heat exchange analytical model

The mathematical task setting for the first stage will include equation (19) with boundary condition (21) as well as the following boundary conditions, with which it should be complied at the temperature perturbation front:

$$\Theta(q_1, Fo) = 0; \tag{23}$$

$$\partial\Theta(q_1,\mathrm{Fo})/\partial\xi = 0; \tag{24}$$

$$(0 < \mathrm{Fo} \le \mathrm{Fo}^*; \quad 0 \le \xi \le q_1(\mathrm{Fo})),$$

where relations (22), (24) represent the coupling conditions for the heated up area and non-heated up area. Relation (23) sets the equality of the body temperature in point $\xi = q_1(\mathrm{Fo})$ to its initial temperature. According to (24), heat flow does not spread beyond the limits of the temperature perturbation front.

It should be noted that at the first process stage, task (19), (21), (23), (24) is absolutely not determined beyond the limits of the temperature perturbation front. In this connection, there is no need to comply with the initial condition of form (20) over the whole thickness (this condition is absent in the mathematical task setting (19), (21), (23), (24)). In this case, compliance with condition (24) is rather sufficient, according to which the body temperature for every $\xi = q_1(\mathrm{Fo})$ is equal to the initial temperature. In addition, task (19), (21), (23), (24) also lacks the boundary condition of form (22) due to the fact that it causes no impact on the heat exchange process at the first stage.

Solution of task (19), (21), (23), (24) is found as the following series:

$$\Theta(\xi,\mathrm{Fo}) = \sum_{k=0}^{n} a_k(q_1)\,\xi^k, \tag{25}$$

where $a_k(q_1)$ − unknown factors. To determine them, let us use boundary (21), (23), (24). By substituting (25) (limited to three series terms) in the listed boundary conditions, to determine $a_k(q_1)\ (k = 0, 1, 2)$, the authors will have the system of three algebraic linear equations. After determining $a_k(q_1)$, relation (25) will be:

$$\Theta(\xi,\mathrm{Fo}) = \mathrm{Ki}(\mathrm{Fo})(0{,}5q_1 - \xi + 0{,}5\xi^2/q_1) \tag{26}$$

where $\mathrm{Ki}(\mathrm{Fo}) = \dfrac{q(t)\delta}{\lambda T_0}$.

To find unknown time function $q_1(\mathrm{Fo})$ in a first approximation, let us compose equation residual (19) and integrate it within the depth of the thermal layer (which is equivalent to making the heat balance integral) of the form:

$$\int_0^{q_1} \frac{\partial\Theta(\xi,\mathrm{Fo})}{\partial\mathrm{Fo}}\,d\xi = \int_0^{q_1} \frac{\partial^2\Theta(\xi,\mathrm{Fo})}{\partial\xi^2}\,d\xi. \tag{27}$$

By calculating the integral in the right part (27), let us find:

$$\int_0^{q_1} \frac{\partial\Theta(\xi,\mathrm{Fo})}{\partial\mathrm{Fo}}\,d\xi = \frac{\partial\Theta(q_1,\mathrm{Fo})}{\partial\xi} - \frac{\partial\Theta(0,\mathrm{Fo})}{\partial\xi}. \tag{28}$$

By considering (21) and (24), relation (28) will be:

$$\int_0^{q_1} \frac{\partial\Theta(\xi,\mathrm{Fo})}{\partial\mathrm{Fo}}\,\mathrm{d}\xi = \mathrm{Ki}(\mathrm{Fo}) \tag{29}$$

By substituting (26) in (29) (assuming $\mathrm{Ki} = \mathrm{const}$), relative to $q_1(\mathrm{Fo})$, let us draw an ordinary differential equation of the form:

$$dq_1^2 = 6d\mathrm{Fo}. \tag{30}$$

By integrating equation (30) in initial condition $q_1(0) = 0$, let us find:

$$q_1 = \sqrt{6\,\mathrm{Fo}}. \tag{31}$$

While assuming $q_1(\mathrm{Fo})$ =1, let us obtain the end time of the first process stage $\mathrm{Fo} = \mathrm{Fo}^* = 0.1667$.

By substituting (31) in (26), let us find the solution of task (19), (21), (23), (24) in a first approximation:

$$\Theta(\xi,\mathrm{Fo}) = 0.5\mathrm{Ki}\left(\sqrt{6\,\mathrm{Fo}} - 2\,\xi + \frac{\xi^2}{\sqrt{6\,\mathrm{Fo}}}\right). \tag{32}$$

The analysis of temperature calculations by formula (32) allows concluding that within the range of the Fourier numbers $0.03 \le \mathrm{Fo} \le \mathrm{Fo}^* = 0.1667$, their difference from the exact solution is 6%, and with $\mathrm{Fo} \le 0.03$ – 1% [6].

Let us find the solution to task (19), (21), (23), (24) if the heat flow is the linear function of time:

$$\mathrm{Ki}(\mathrm{Fo}) = \mathrm{Ki}\ \mathrm{Fo}. \tag{33}$$

The differential equation relative to $q_1(\mathrm{Fo})$ will be here:

$$\frac{dq_1^2}{d\mathrm{Fo}} + \frac{q_1^2}{\mathrm{Fo}} = 6. \tag{34}$$

By integrating equation (34), in the initial condition $q_1(0) = 0$, let us find:

$$q_1(\mathrm{Fo}) = \sqrt{3\mathrm{Fo}} \tag{35}$$

Formulas (26), (35) represent solution to task (19), (21), (23), (24) in a first approximation (at linear change of the heat flow over time).

The unidimensionality of the boundary problem for the first stage of the heat conductance process is conditioned by the fact that surfaces of items which thicknesses are negligible as compared to the cross dimensions of the irradiation spot subjected to millisecond pulse laser irradiation. In this connection, the heat flow value in the irradiation spot surface may be neglected [10 – 13].

For the sake of convenience of calculating the thermal condition of particular materials, let us represent formula (32) in the dimensional form:

$$T = T_0 + 0.5\frac{q(t)\delta}{\lambda}\left[\sqrt{6\frac{at}{\delta^2}} - 2\frac{x}{\delta} + \frac{(x/\delta)^2}{\sqrt{6at/\delta^2}}\right], \qquad (36)$$

where $q(t) = q_0 \frac{at}{\delta^2}$.

Formula (36) for the plate surface $(x = 0)$ will be:

$$T(0,\, t) = T_0 + 0.5\frac{q(t)}{\lambda}\sqrt{6at}. \qquad (37)$$

The results of temperature calculation by formula (37) for aluminium are given in fig. 7 ($\lambda = 204$ *W/(m·K)*; $a = 91\cdot10^{-6} m^2/s$; $T_0 = 20°C$). It follows from their analysis that the plate surface flatness increases without limit over time. However, in many practical cases it is required that the surface temperature, after having reached some set value over some time, would further remain constant over time. Let us note that the surface temperature invariability may be ensured only until the temperature perturbation front reaches the opposite wall of the plate. After it reaches coordinate $(x = \delta)$, the whole plate will participate in the heat exchange process, and reaching the constant wall temperature over time will not be possible anymore - it will infinitely rise over time. Therefore, reaching the time-constant temperature is only possible at the first stage of the process. To this end, it is necessary, by using the formula (37), to find such regularity for the change of the heat flow over time $q(t)$ to make the plate surface temperature $(x = 0)$ set and invariable over time. By expressing $q(t)$ from (37), let us obtain:

$$q(t) = \frac{2\lambda(T(0,t) - T_0)}{\sqrt{6at}} \quad (t > 0) \qquad (38)$$

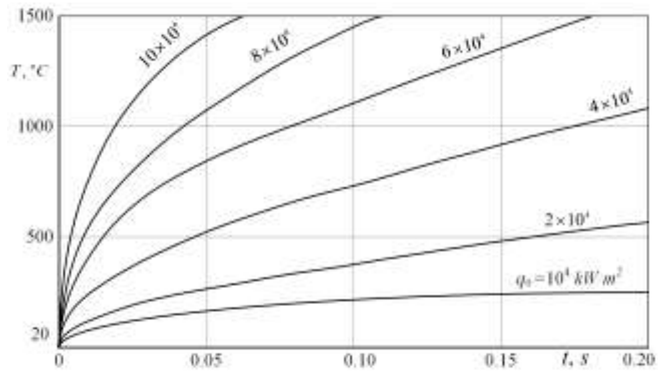

Fig. 7 Temperature change on the surface of the aluminium plate depending on the laser irradiation flow

According to formula (38), by setting any particular plate surface temperatures, the respective formulas may be found for heat flow change. The change of laser flow over time should be arranged in accordance with these formulas. Heat flow-time curves for different temperature values on the aluminium plate surface are given in fig. 8. By analysing the obtained results, it may be concluded that to achieve any set temperature of the item surface, the heat flow is reduced at the insignificant section of the initial time range $(t \neq 0)$ from the maximum value at the initial time moment to some minimum value, which is further practically time-invariant.

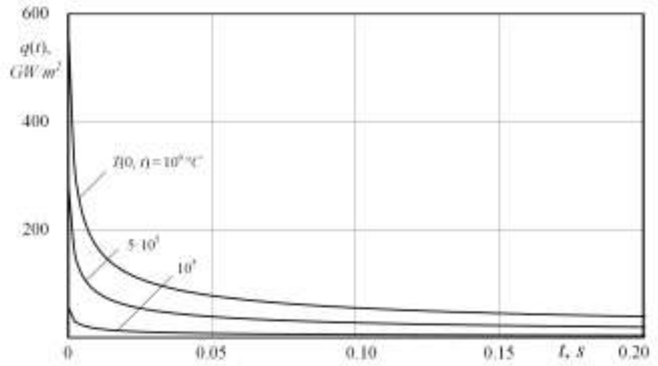

Fig. 8 Change of heat flow over time (as per formula (38))

VI. Conclusion

1. The mathematical model of local non-equilibrium heat exchange in a plate with external pulse heat source was developed allowing one to imitate the process of high-frequency laser irradiation of materials, considering the relaxation properties.

2. It was shown that consideration of relaxation processing of materials results in delayed heating of the item at the initial time range due to the physical impossibility and immediate setting of boundary conditions (of the first-, second- and third class), connected with resistance of the medium material to the temperature change process.

3. Within the time range of the first stage of the process, when the temperature perturbation front has not yet reached the opposite wall of the plate, the formula for change of the heat flow over time was obtained allowing maintaining the set temperature at the wall $(x = 0)$ for any particular material.

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