Research and modeling of laser ablation by ultra-short laser pulses for metal targets

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Abstract. In this article a new mathematical model for a laser metal ablation by ultra-short duration laser pulses is proposed. The computational process is based on a two-temperature hydrodynamic model for electrons and ions. Some improvements are made and wide-range equation of state for metals is included in model. The results of the computer simulation of ablation depth for aluminum and copper are compared with experimental data at a different laser fluency values. We obtained a good agreement between the experimental and calculated data on the ablation depth.

1. Introduction

Currently, lasers are widely used in various technical applications: metal cutting and welding [1, 2], nanotechnology [3-5], medicine [6-9], chemistry [10], and many other fields [11-17]. The effect of laser radiation on substances and even on individual classes of substances (for example, metals) turned out to be very complex in terms of the size of the various physical processes accompanying it [18, 19]. Of course, as a result of many theoretical and experimental studies, regularities of such processes have now been established, but a number of important points remain not fully disclosed.

Now for research in the field of laser technology is actively used mathematical modeling of physical processes. The formulation of computational experiments based on experimental studies made it possible to carry out complex and detailed studies with much lower material costs. This we owe a significant increase in computing power in the past few decades, the emergence and development of parallel computing and supercomputer technologies [20, 21]. The most important element of modeling physical processes is the development of a mathematical model that adequately describes the processes in question. There are two main approaches to creating such models: discrete and continual.

In the continual approach, systems of partial differential equations of continuum mechanics are used to describe processes. They describe the macroscopic characteristics: temporal and spatial distributions of electromagnetic, thermal, and other fields. In the discrete approach, processes at the micro level are described. For example, in the molecular dynamics, the motion of a set of particles (molecules, atoms, ions) with a selected interparticle interaction potential is considered and the corresponding equations of motion are integrated at each time instant to determine the position of the particles in space. Averaging the found coordinates and the found momenta of the particles, we can
study in general the dynamics of the occurring physical processes [22]. An important advantage here is that no (or just few) further assumptions on the processes are required for calculations, but simulation requires a lot of computational resources, which significantly limits the size of the study area or time frame of the processes under research [23].

In the continual approach, on the contrary, a relatively small amount of information is needed, and the calculations are carried out with average values of physical parameters located on an infinitely small amount. At thermodynamic equilibrium, physical characteristics can be determined in various ways: experimentally, by counting from distribution functions (Fermi for electron gas, Maxwell-Boltzmann for ideal plasma or ideal gas, Bose for phonon gas and others). If thermodynamic equilibrium is locally violated, then distribution functions can be found by solving kinetic or quantum-kinetic equations. Actually, the presence of a certain distribution function is the information that can be used to describe non-equilibrium physical processes with good accuracy. However, there are a number of problems, such as processes associated with fast phase transitions, where the use of a discrete approach can give very useful results.

When exposed to high-power laser radiation on metals, thermodynamic equilibrium is disturbed. Laser radiation is absorbed primarily by electrons, then transferring energy to ions, which, because of their massiveness, change their temperature more slowly. As a result, physical processes are described in the two-temperature approximation [24], where each subsystem has its own temperature and equations of state. Here, an important problem is the determination of various physical characteristics that vary over a wide range in a short time. For the metals and ultra-short duration laser pulse such physical processes and their approximate time scales are shown in Figure 1.

![Figure 1. Processes and approximate time scales for metal ablation by about 100 fs duration laser pulse.](image)

Laser pulse ablation is the process of removing a substance after exposure to high-intensity laser pulses. Pulses with a shorter duration (femtosecond or picosecond) are preferred for various applications, because the ultrashort pulse duration limits thermal diffusion, which improves the quality of processing. With the help of exposure to femtosecond laser radiation obtained the most high-quality and highly reproducible results of material processing [25]. The interaction between target material and the laser beam will result in the ablation of the target and consequently the formation of a crater on the target surface [26].

There are many mathematical models used to simulate ablation processes, where the two-temperature model (TTM) is commonly used. In this model the energy transfer in the metal is described by two coupled generalized equations of heat conduction (with different the electrons and the lattice temperatures). This model has received significant development over the past decades such as dual-parabolic two-step model, semiclassical two-temperature model, etc. [27]. Also coupling TMM with molecular dynamics method (TTM-MD) or with classical heat accumulation model is often used for creation hybrid models [28, 29].

The material removal processes in this model are often described by a system of hydrodynamic equations (TTM-HD). However, when interaction between material and laser radiation is studied, an appropriate calculation for the thermodynamic material properties is required over a wide region of states including normal condition or even plasma state at high pressures and temperatures [30, 31].
Therefore, there is still a need to develop a mathematical model for sufficiently quickly and enough accurately computer simulation of laser ablation [32, 33]. In our article we propose an improved mathematical model in two temperature form with using the developed wide-range two-temperature equation of state for metals.

2. Mathematical model

2.1. System of equations

The problem of irradiation with an ultra-short laser pulse of a metal target is considered. The mathematical model for ablation in one-dimensional case is proposed in the two-temperature form based on conservation laws for the mass, the momentum and the energy of electron and ion subparts:

\[
\frac{\partial}{\partial t} \left( \frac{1}{\rho} \right) + \frac{\partial \nu}{\partial m} = 0
\]  
\[
\frac{\partial \nu}{\partial t} + \frac{\partial P}{\partial m} = 0
\]  
\[
\frac{\partial e_e}{\partial t} + P_e \frac{\partial \nu}{\partial m} = - \frac{\alpha_{ei}}{\rho} (T_e - T_i) + \frac{\partial}{\partial m} \left( k_P \frac{\partial T_e}{\partial m} \right) + I
\]  
\[
\frac{\partial e_i}{\partial t} + P_i \frac{\partial \nu}{\partial m} = \frac{\alpha_{ei}}{\rho} (T_e - T_i)
\]  

where \( m = \int_{x=0}^{x} \rho dx \) – is the mass coordinate, \( x(m,t) \) – particle trajectory with coordinate \( m, \rho \) – is the density, \( \nu \) – is the velocity, \( t \) – is the time, \( T_e \) and \( T_i \), \( e_e \) and \( e_i \), \( P_e \) and \( P_i \) - the temperatures, internal energies and pressures of electrons and ions, \( P = P_e + P_i \) and \( \varepsilon = e_e + e_i \) - the sums of the pressure and the internal energy, \( \alpha_{ei} \) - electron-ion relaxation coefficient, \( I \) - absorbed laser radiation energy, given by formula based on Beer–Lambert law:

\[
I = \frac{B}{\tau_L \sqrt{2\pi} e} \exp \left( \frac{x(m_0,t) - x(m,t)}{\delta} \right) \exp \left( - \frac{t^2}{\tau_L^2} \right)
\]

where \( B \) is the laser radiation energy, \( \tau_L \) - the duration of the laser pulse, \( \delta \) - the metal skin depth, \( m_0 \) – the surface coordinate of metal target.

Initial conditions are the density of metal at room temperature \( T_0 \), the room temperature and velocity equal to zero at zero time. The boundary conditions are zero pressure and zero heat flux through the boundaries:

\[
\rho|_{t=0} = \rho_0, \quad T_e|_{t=0} = T_i|_{t=0} = T_0, \quad \nu|_{t=0} = 0
\]  
\[
P_L = P_R = 0, \quad \frac{\partial T_e}{\partial m} |_{L} = \frac{\partial T_i}{\partial m} |_{R} = 0
\]

The electron wide-range thermal conductivity coefficient \( k \) is constructed as an interpolation between limiting metal and hot plasma case according to the Drude formula:

\[
k = \frac{c_e \left( \frac{6 k_F}{m_e} \right)^2 + \left( \frac{3 k_B T_e}{m_e} \right)}{3 v_c}
\]

where \( c_e \) is the electronic heat capacity, \( E_F \) is the Fermi energy, \( k_B \) is the Boltzmann constant, \( m_e \) is the electron mass, \( v_c \) is the collision frequency, which is stitched from \( v_{ee}, v_{eF}, v_{ei}, v_{ii} \), where the indices \( ee \) and \( ei \) refer to electron-electron and electron-ion collisions, and \( pl \) and \( c \) to plasma and condensed matter [34]:

\[
\nu_c = \left( \nu_{ee}^{-2} + \nu_{eF}^{-2} \right)^{-\frac{1}{2}} + \left( \nu_{ei}^{-2} + \nu_{ii}^{-2} \right)^{-\frac{1}{2}}
\]

The system of equations is complemented by the equation of state, which will be discussed below.

2.2. Wide-range equation of state for metals

To solve the system of hydrodynamic equations in proposed mathematical model we supplement it with equation of state for metals in two-temperature form, where to describe the thermodynamic properties of a metal (pressure, internal energy and temperature) the free energy \( F \) is used. The atomic cell volume \( V \) and temperatures \( T_e, T_i \) are used as the main thermodynamic parameters. The basic assumption on which the described model is based is that, for any density and temperature, all electrons can be divided into free and bound. Strictly speaking, this separation has a clear physical
meaning only in the case of an ideal plasma, but it will be preserved in the area where the effects of nonideality play a fundamental role, assuming that the contribution of these effects to pressure and energy is adequately described by $F_{el}$. The number of free electrons in a single atomic cell is called the degree of ionization and denoted by the letter $y$.

For a numeric cell it is described as sum of the electronic and ionic components, and the part responsible for the interaction between them:

$$F = F_e + F_{i} + F_{el}$$

(10)

Cell volume $V$ and temperatures $T_e$, $T_i$ are used as main thermodynamic parameters. The electron pressure is described as the ideal Fermi electron gas pressure with temperature $T_e$ and density $\frac{y}{V}$, where $y$ is received by solving the slightly modified equation of ionization:

$$\mu_F\left(\frac{y}{V}, T_e\right) + I(y) - b\left(\frac{y}{V}\right)^\beta (1 + \mu T_e V^\sigma)^{-1} = 0$$

(11)

here $\mu_F$ - the Fermi gas chemical potential, $I(y)$ - ionization potential, which is appear in the equation as a smoothing spline by the experimental stages of ionization values - $I(1)$, $I(2)$ etc. $B(V, T_e)$ is used to take into account cold ionization of compressed matter and $b$, $\beta$, $\sigma$, $\mu$ - parameters, which are constructed using shock compression experimental data for metals.

For the describing free energy we use approximation of an ideal Fermi gas:

$$F_e = y T_e \left(\frac{3}{2} \varphi - \frac{3}{2} \ln \left(1 + \frac{5}{2} \varphi\right)\right)$$

(12)

Second part $F_{i}$ is linked to the transition from states at different temperatures $[35]$:

$$F_i = \frac{3}{2} T_i \ln \frac{1 + \lambda G}{1 + \lambda \varphi}$$

(13)

where $G = \frac{4\pi^3}{3} \frac{y_c(V)}{V^3}$ and $y_c(V)$ is a function of the cell volume $V$, which is defined by the ionization equation. Using Helmholtz free energy, we receive necessary formulas for the pressure of the electrons, the internal energy and chemical potential:

$$P_e = \frac{1}{5} (3\pi^2)^{\frac{2}{3}} \left(\frac{y}{V}\right)^{\frac{5}{3}} + T_e \left(\frac{y}{V}\right)^\beta \left(1 + \frac{2}{5} \varphi\right)^{-1}$$

(14)

$$\varphi_e = \frac{3}{2} V P_e$$

(15)

$$P_i = \frac{T_i}{V} \ln \frac{1 + \lambda G}{1 + \lambda \varphi}$$

(16)

$$\varphi_i = \frac{3}{2} T_i \ln \frac{1 + \lambda G}{1 + \lambda \varphi}$$

(17)

$$\mu_F = \frac{1}{2} (3\pi^2)^{\frac{2}{3}} \left(\frac{y}{V}\right)^{\frac{5}{3}} + T_e \left(1 + \frac{2}{5} \varphi\right)^{-1} \ln \left(1 + \frac{5}{2} \varphi\right)$$

(18)

And finally, in $P_{el}$ we consider that it should not have an influence on the equation of ionization, total pressure at normal density and zero temperature must be equal zero, at a fixed density interaction between electrons and ions should decrease as the temperature increases, at low densities it must decrease fast:

$$P_{el} = -\frac{1}{5} (3\pi^2)^{\frac{2}{3}} \left(\frac{y}{V_0}\right)^{\frac{5}{3}} \left(\frac{\varphi}{\varphi_0}\right)^\delta \left(1 + \frac{\delta}{1 + (\delta + 1) \varphi / T^*}\left(\frac{\varphi}{\varphi_0}\right)^\delta\right)$$

(19)

where $V_0$, $\gamma$, $\delta$, $T^*$ are the parameters, which can be configured using values of the density, of the sublimation energy, of the isothermal compressibility and of the metal thermal expansion coefficient.

The solution of the system of equations of the whole mathematical model is carried out by splitting into physical processes; we consider separately the hydrodynamic motion of a substance during the absorption of laser radiation, the electronic thermal conductivity and the energy exchange between electrons and ions. To calculate the ablation depth, we perform an integration of the mass flux through the surface:

$$d = \frac{1}{\rho_o} \int_0^t \rho V |_{x=x_0} dt$$

(20)

3. Results and discussion

To check the developed equation of state, coefficients are calculated, and comparisons are made with various experimental data. As an example, Figure 2 shows the shock adiabat of aluminum.
Figure 2. The calculated shock adiabat of aluminum and experimental data on it.

According to the Figure 2, we can conclude that the equation of state describes the behavior of the metal quite well in a wide range of pressures and densities.

Figure 3. The calculated laser ablation depth for aluminum and experimental data on it.

Figure 4. The calculated laser ablation depth for copper and experimental data on it.

To verify our mathematical model, the ablation depth for aluminum and copper is calculated and compared with the experiment. This experimental data is collected for an average depth of machined grooves after using laser pulses with 170 fs duration. In Figure 3 and 4 the results are shown.
The results agree well with experimental data for different values of the laser fluence. For large fluence values, the estimated crater depth is slightly overestimated, due to the fact that in proposed improved TTM model the effects of plasma shielding are not considered now [36].

For copper we also compare results with experiment values of ablation depth for different values of the laser fluence which are received by multiple laser pulses with 100 fs duration with same total energy (Figure 5).

![Figure 5. The calculated multi-pulse laser ablation depth for copper. 1, 2 – experimental data, 3 – computer simulation results.](image)

In general, the model can be employed for computer simulations of such processes, however, there is some inconsistency with the experiment. It can be explained by the fact that the experimental ablation depth per pulse is estimated in the way that the measured groove depth is divided by the number of laser shots. This multi pulse laser radiation in the same spot may lead to the changes in the surface optical properties because of increasing roughness. Furthermore, inclined groove walls in the case of deep groves could significantly change the reflection of a laser beam. Some investigations of these problems are needed, and the results may be included in this model later.

4. Conclusion
An equation of state has been developed to simulate the effects of ultra-short laser studies on metals. Using it in a two-temperature improved mathematical model, computer simulation of the ablation depth for various metals was performed. A fairly good agreement with experimental data under various conditions was obtained. Possible further ways to improve the model are noted.

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