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CONTEMPORARY PROBLEMS, EXPERIMENTS, THEORY, PHASE TRANSITIONS PROVOKED BY LASER BEAMS IN CONDENSED MATTER

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ABSTRACT: *This paper analyses the broad issues interaction of laser beams with condensed material. For purpose of lasers in materials processing, many of the fundamental and phenomenological models were developed of which the authors have chosen some and rated the results of the interaction of selected examples. Examples are based on the possible existing types of lasers and operating modes, the energy and pulse width. For given real pulses, the distribution of temperature is estimated for two models of time and phase transitions. The interaction is considered for Dirack δ-pulse and the pulse of complex shape. For given operating parameters, experiments were performed, which have shown the possible further analysis of the interaction, which do not offer thermal models in the first place. Times when it comes to the melting were estimated using applied model and results show that predictions, based on this model, are much better for metals like steel than for aluminum and copper, which were problematic for treatment from the beginning of laser technique.*
KEYWORDS: *laser beam effects, numerical simulation, laser material interaction*

INTRODUCTION

Having in mind large energy range (energy density) and working temporal duration of quantum generators, rich experimental experience, industrial solved technological operations, etc., those will reflected to some chosen topics, which are linked (it seems) to some unsolved fundamental questions. Experimental facts could be and should be with possible interpretations based on microscopies (light and electronic- SEM), profilometry, X ray investigations, microhardness, etc. Special attentions will be paid to modeling of temperature distribution in exposed metallic samples as well as modeling of some other processes induced at irradiations when chosen equation is inappropriate.

The process of oxidation linked with laser interaction can be presented in material by simple model linked to elemental content (in steel it can be presented by simple model linked to carbon content). Nucleation processes, phase transitions, and relationship with critical phenomena are interesting approaches, beside unavoidable nonlinear phenomena as well [1-5]. Laser interaction phenomena can be treated as one of beam techniques (neutron, gamma, ions, electron)[6]. Interesting topics can be linked to different transition amorphous to crystal and vice versa, in common phases (α,β,γ,δ) in iron (steel) transformations and other respective processes. Nucleation processes and relationship with critical phenomena [7] are linked with various regimes of material expositions of steel, aluminum and other element based materials and experimentally realized. Full attention has to be paid to linear and nonlinear parts of reflection, absorption as well as thermodynamic parameters (conductivity, specific heat coefficient etc.) [8].

Sintering and powder metallurgy have close links considering technological-metallurgical processes: new aspects are that the powder for „classic“ -oven sintering processes can be obtained by laser techniques and it can be characterized by new specific distribution functions by particle size and other quantitive properties of powder (monodispersity, etc.)[9].

Considering laser working regimes, first laser techniques for powder obtaining are characterized by larger pulses (Nd^{3+}: YAG, Q switch regimes). New methods include fem to second laser systems. Powder metallurgy has been obtained for new categories. Due to the links between mechanical, thermal, acoustics and other features with optical characteristics, it has to be paid attention that new powder have to have new magnitudes of electrical resistance, absorbance etc., especially in the relationship of linear and nonlinear materials behavior.

The unsolved tasks are in areas of: interpretations of nonlinear data for coefficient of reflection and methods with complex measuring schemes with temporal resolution, where exist laser beam for *testing and working interaction beam, which can transform material phases [8,10]. Optical features can be considered through quantum mechanical approaches, Maxwell equations, polarizations formalisms, matrix calculus, etc. Powder characterization and light (laser beam scattering) on the other hand give the other point of view, considering white light scattering, and changing of powder color. Very impressive color changing can be followed by milling processes and changing the powder characteristic sizes [11-14].*

Microhardness changes at local or on larger area (obtained by scanning processes) are complex; depending on the working regime, scanning rates, laser power density.

Results are linked also to measuring-testing systems, for plate density measuring and, depth controls, following wavelness, etc.[15]. For nonferrous metals there are indicions that microhardness could be increased as well as decreased. Our experiments with free generation regime have shown the dispersion of results. The obtained mechanical stresses and acoustical processes, annealing, have a lot of opposites (inverse) provoked processes which describe laser-material interactions [16,17]. Here are Raman spectra as modern measuring method (Linewidth and line positions of spectra changes provocated by mechanical stresses [18]. The Zeeman effects are also linked to the stresses. A lot of electrooptical and paramagnetic -optoacoustic effects are induced and can be found further. Very sophisticated theories exist, as well as some for rule of thumb evaluations, which are very simple but limited.

The link with surface tensions, and mechanical moduli, sound velocity and induced Brillouin scattering, the evaluation of transitions amorphe-crystal states and vice versa, the links with laser wavelength and interference fringes -morphology structures modulated through laser interactions, the links with energy of fractures through the limiting energies (power) which follow the self-focusing are unavoidable topics for transparent and nontransparent materials. Thin films have multifunctional role: various materials obtained by specific techniques, including laser depositions, surface changes features by irradiations, induced reactions. Depending on the pulse duration (or pulse length), laser interaction with material could be treated as relatively new techniques for dynamics of electron ensembles, etc. Here are also: crystallization, modeling of various complexity degree, different approaches to thermal distribution modeling, etc.

The simulation of laser interaction with material is analyzed for some materials of metallic and semiconducter types by various models. Some of them will be presented in this paper. At the end we are presented some of our micrographs to illustrate of complexity in the modeling of laser material interaction and reality.

THE SIMPLE THERMAL APPROACH

One approach by general thermal equation uses only some part of equation which general solution doesn't exist. The model uses pulses of Dirack-like shape. Maximal energy of Dirack pulse is chosen as 100 mJ. The laser spot is regularly circular shape 2mm dia. Laser beam intensity at maximal energy for 2mm dia, is $31.83 \bullet 10^3$ J/m^2. For temperature distribution in time and space, during the exposition of material, heat equation has to be solved. For this approach, boundary conditions for start and final „points" have to be precisely determined. Initial conditions are T(z,0)=f(z), where f(z) functions, which describe temperature distribution in the points z, for t=0. In our case, the T_0 is a room temperature T_0=20^{o}C (293K). First boundary condition is (1):

$$I(z)\big|_{z\to 0} = -\lambda \frac{\partial T}{\partial z}\big|_{z\to 0} \Rightarrow T_{z\to 0} = -\frac{1}{\lambda}\int I(z)dz \qquad (1)$$

Dependence of intensity versus depth is described by Lambert-Beer law. The data for simulation, for chosen materials, are presented in Table 1 as well as needed parameters.

Table1. Properties for chosen materials

Material	ρ-density (kg/m^3)	Cp-specific heat (kJ/kgK)	α-Temp. difusivity*
Steel 304	$7.93*10^3$	0.5	0.005397
Al	$2.3*10^3$	0.9	0.001
Si	$2.33*10^3$	0.7	0.026

** this and other needed parameters are normalized*

a) For stainless steel 304: parameter α=0.05. With adequate I0: $T_{z=0}$=365.69 ^{o}C=638.84K is obtained.

b) For Al: parametar α =0.001, R=0.91. With adequate I_0 : $T_{z=0}$=298,14 ^{o}C=571.3K is obtained.

c) For Si: A =0.001, α=0.026. With supposed value for I0 we obtain: $T_{z=0}$=1748.42 K.

*The solution of the respective equation can be performed in Matlab 7.5, M-files, where we created interface program **toplota.m**. With k, in heat function (**toplota(k, z, t, inite, bdry)**), is normalized thermal diffusivity (our case for k=0.005397, i.e.. k=0.001, and k=0.026 for steel, i.e. Al and Si, respectively.). The created M-file is used for 1D solution of thermal equation in temporal intervals [0, 1s], with Δt=0.01s, up to the depth of 1 mm with resolution Δz=0.01 mm. All parameters used in this program are normalized. The solutions in graphical forms are presented in Figures 1, 2, 3 for stainless steel, aluminum and silicon on respective way.*

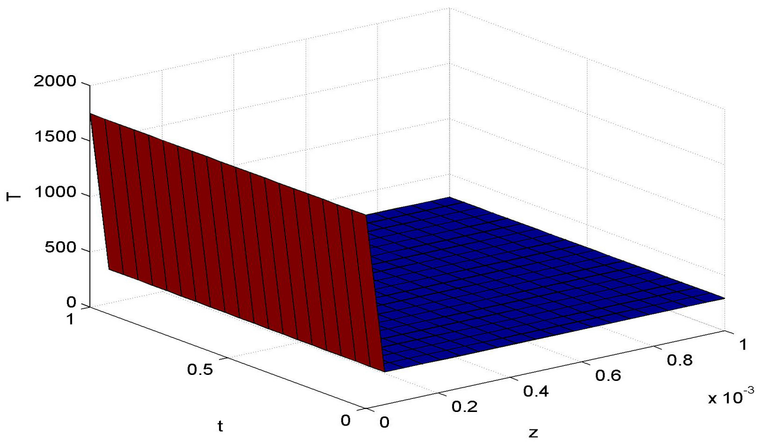


Figure 1. Temperature distribution in 304 stainless steels after laser pulse: pulse energy 100 mJ, the pulse width is considered as neglecting parameters. The state is up to the depth of 1mm, 1s after laser expositions

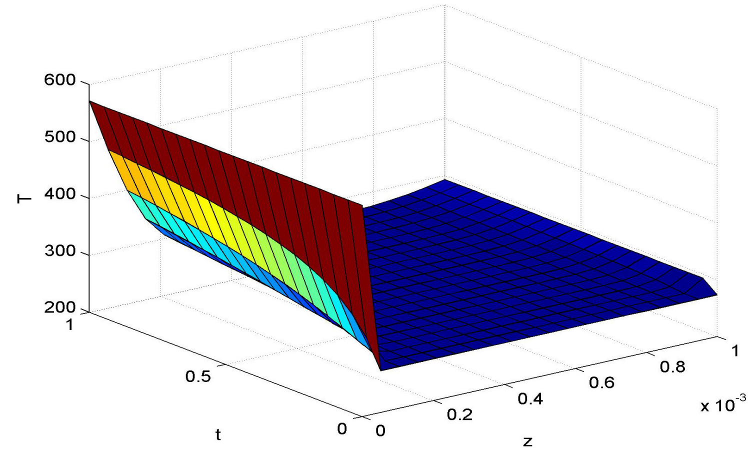


Figure 2. Temperature distributions in aluminum Al after laser pulse: pulse energy 100 mJ, the pulse durety is considered as neglecting parameters. The state is up to 1mm depth, 1s after laser expositions

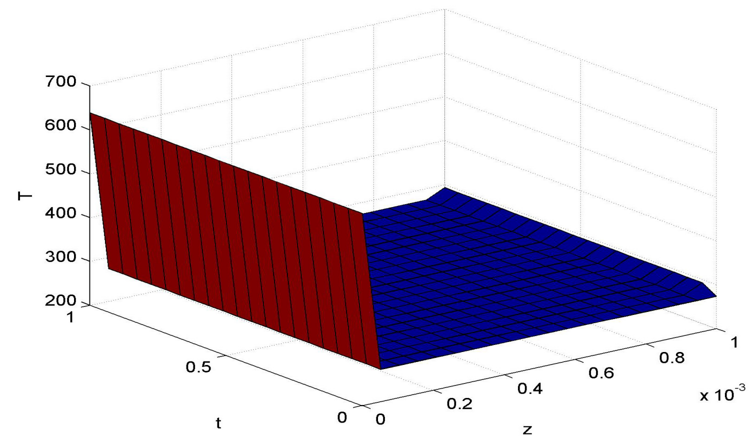


Figure 3. Temperature distribution in Si after laser pulse: pulse energy 100 mJ, the pulse durety is considered as neglecting parameters. The state is up to 1mm depth, 1s after laser expositions

OTHER SIMPLER THERMAL APPROACHES

The initial equations based on heat model and some results of our calculation and from references we analyzed and evaluated from the treatment of general heat equation, considering the material from several point of view.

The approach is carried out having in mind expectations of users and what really occurred during laser interactions with solid state [19-24].

The total enthalpy approach

Consideration starts from the thermal equations [21,22,25,26].

$$\frac{\partial}{\partial t}(\rho H) = \nabla(k\nabla T) + S \qquad (2)$$

where ρ is the material density,
T-temperature,
k-thermal conductivity,
H-enthalpy and
S-function describing local heat generation during an interaction.

S contains terms determined by laser working parameters, which could be modeled either in a cw or various pulsed working regimes. A frequent approximation for Nd^{3+}: YAG laser pulse is triangle pulse. In this paper more sophisticated approximation relative to real pulse is used (Figure 4).

Common assumption used for the evaluation of (2) are: one dimensional case density independent on temperature, time or phase state. This latest assumption is problematic, since density is a function of all mentioned variables and generally three dimensional considerations are often necessary. In most cases is practical interest a solution of S [25, 26] can be expressed as (3)

$$S(x,t)=(1-R(x,t))P(t)=P(t)\alpha e^{-\alpha x} \qquad (3)$$

where R is the coefficient of reflections, α- coefficient of extinction and P-the power density of laser pulses. More complex solutions can be found in [27,28].

By the method of numerical discretization, a classical model of crystal growth, based on heating and nucleation can be included in the consideration. The solutions of (2) and (3) could be obtained from (4):

$$\rho\frac{H_i^{n+1}-H_i^n}{\Delta t} = \frac{1}{(\Delta x)^2}\left[\frac{k_{i+1}+k_i}{2}\left(T_{i+1}^n - T_i^n\right) + \frac{k_i+k_{i-1}}{2}\left(T_{n-1}^n - T_i^n\right)\right] + S_i^n \qquad (4)$$

This approach has been successfully used for many materials Si, HgCd (Te), etc.

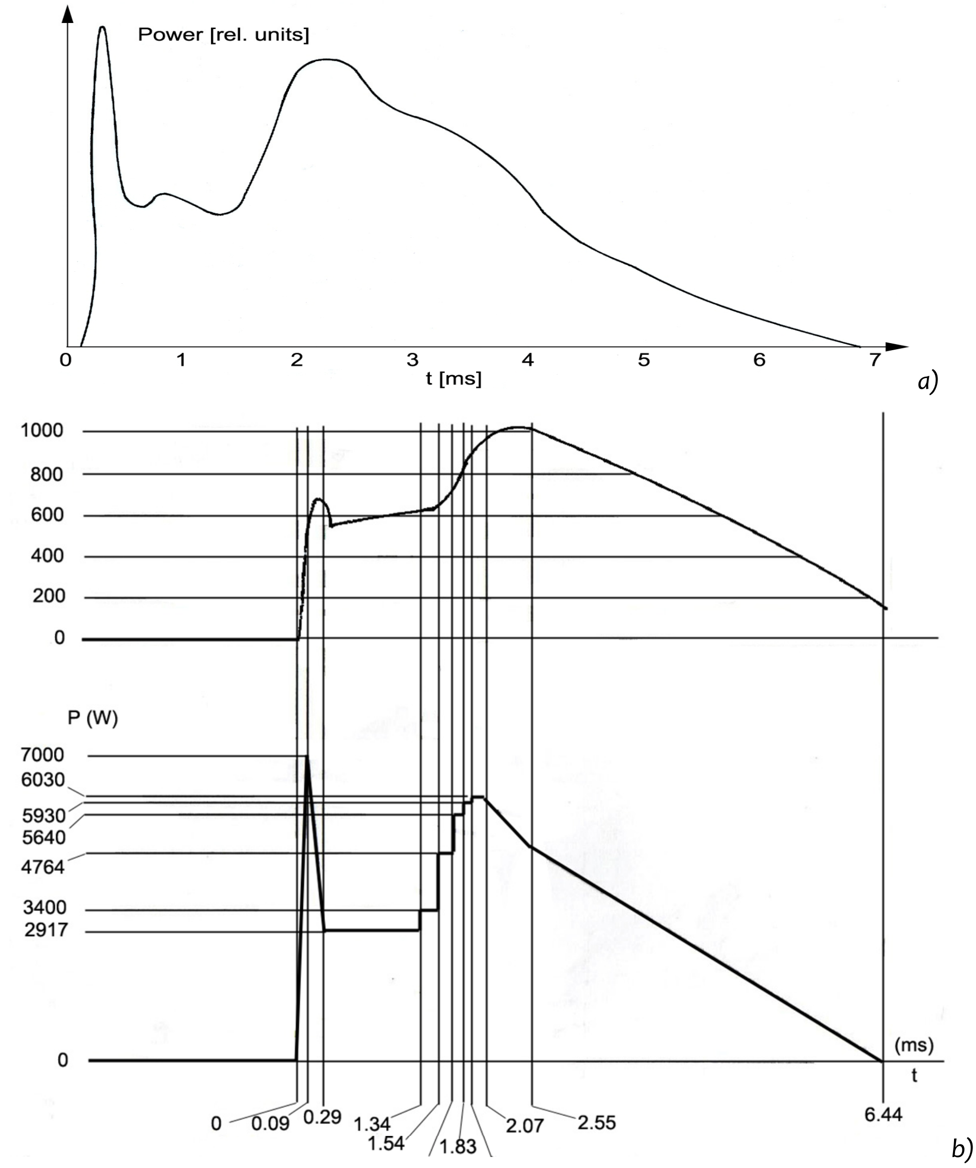


Figure 4. Real shape of pulse Nd^{3+}:YAG laser a) and detail of division for further analyses b).

Melted front propagation approach

a) Constant beam intensity approximation (constant parameters).

To simplify the solution the heat conduction is divided in two parts, that corresponds to solid (s) and liquid (l) state, so two separate equations (5, 6) exist.

This is based on the depth of penetration and the speed of the laser-melted front calculation, from the analytic solution of thermal equations of the similar forms as (2):

$$c^{(L)}\frac{\partial}{\partial t}T_L = k^{(L)}\frac{\partial^2}{\partial t^2}T_L + A(x) \tag{5}$$

$$c^{(S)}\frac{\partial}{\partial t}T_S = k^{(S)}\frac{\partial^2}{\partial t^2}T_S + A(x) \tag{6}$$

where $c^{(L)}$ and $c^{(S)}$ are heat capacities per volume for liquid and solid states, and A(x) is an integral heat source of intensity [26]. Term A(x) correspond to term S in equation (2). A rigorous mathematical treatment leads to the expression for velocity of melted front given in equation (7):

$$V = \frac{2P}{m} - \frac{A`Pt^{1/2}}{m^2} - \frac{B`}{Pt^{1/2}} + \frac{B``}{2Pt^{1/2}} - \frac{2P}{m}\exp\left(\frac{-2P\alpha t}{m}\right) \tag{7}$$

where:

$$A` = I_0\frac{(1-R)}{k^{(L)}}\sqrt{\frac{k^{(L)}}{\pi}} \quad B` = 2I_0\frac{(1-R)}{c^{(L)}}\sqrt{\frac{k^{(L)}}{\pi}} \quad B`` = \frac{k^{(L)}T_m}{c^{(L)}} \quad k^{(L)} = \frac{k^{(L)}}{c^{(L)}} \quad m = \frac{\rho L_f}{c^{(L)}}$$

L_f is latent fusion heat, T_m is melting point.

b) Short pulse Gaussian approximation (constant parameters).

The entire procedure and expressions are more complex but the final solutions is similar to (2.6). With iterative Pickard approach more complicated formulae obtains, with new constants and explicit relation dependent on the radius W_0 at which the intensity has fallen to 1/e [28].

c) Constant beam intensity (nonconstant thermal parameters)

All coefficients are dependent on time and space, the density depends on temperature as ~ 1/T. For the front velocity V, a five- term expression (7) is arrived at (8):

$$V = \frac{2P}{m} - \frac{A`P}{m^2 t^{1/2}} - \frac{B`}{Pt} - \frac{B``}{Pt^{3/2}} - \frac{P}{m}\exp\left(\frac{-P\alpha t}{m}\right) \quad (8)$$

whose details are in [25]. Considering a short pulses Gaussian beam in nonconstant parameters approximation seven-term expression [25].

Approximations 1) and 2) lead to quite different results so the approximation with ρ as a constant is justified for small power densities which is not the case for laser interactions, where power densities are large enough to provoke temporal and /or temperature variations of optical and thermal coefficients. Thus the detail knowledge of temperature behavior of constants is necessary for considerations, especially for new and poorly investigated materials. In the case of laser interaction with the layered material, another theoretical approach should be explored verification of the results [29-31]. We made some calculation based on theoretical models mentioned above.

Chosen approach

Some calculations based on theoretical treatment of the heat equation are made, but with another approach with numerical simulation given in part 1 of this paper. For pure metal specimens e.g. Cu, Al and steel are made calculations.

Calculations are performed for pure metal plates using the heat equations (2, 4) and temperature coefficients in the solutions velocity of melted front from the heat transfer equation. Dependence of specific heat is taken in the form of $c=a+bT-cT^2$ and data for the other coefficients in specific temperature ranges differ for the solid and liquid phases.

The real laser pulse (Figure 4) is simplified and divided in sections and the depth of penetration is estimated using the approximation for semiinfinite specimen, taking the relation between the depth of penetration and the specimen thickness, in account. A suitable approximation for the material spectral emmitance of material is incorporated, too. On this way we calculated the time when will be obtained melting process for the most known metals (Table 2). Discrepancies founded for Cu, Al, and steel between the experiment and calculations can be of interest for further investigation.

Table 2. Temperature calculation for the materials exposed to Nd^{3+}:YAG laser with pulse shape of Figure 4 obtained by one of the approximation based on heat equation in time.

τ μs / T^0C	10	20	30	40	50	60	70	80	90
Al	41	70	127	184	248	317	392	472	550
Steel	72	167	290	435	600	782	980	1192	1418

SOME EXPERIMENTAL RESULTS OF DAMAGES AND MORPHOLOGY

In Figures 5-9 we presented some of our experimental results without the details as the presentations of the damage shapes and morphology which can not be obtained through such approaches as we presented. The influence of polarizations, type of materials, etc., can provoke induced structures, surface modifications which have to be explained through other approaches on the theoretical or phenomenological basis.

Figure 5. Nd^{3+}:YAG laser damage of 304 steel specimen (λ=1.06μm, top hat, τ=2x2ms, spot size 2mm, E=9.06J) analyzed by light microscope, 50x

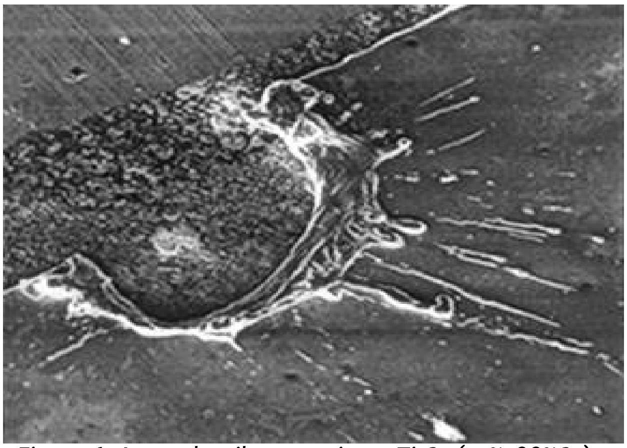


Figure 6. Amorphe ribon specimen Ti-Co (72%- 88%Co); exposed to ruby laser (λ=0.69μm, τ=30ns, E=0.75J) analyzed by SEM, 50x

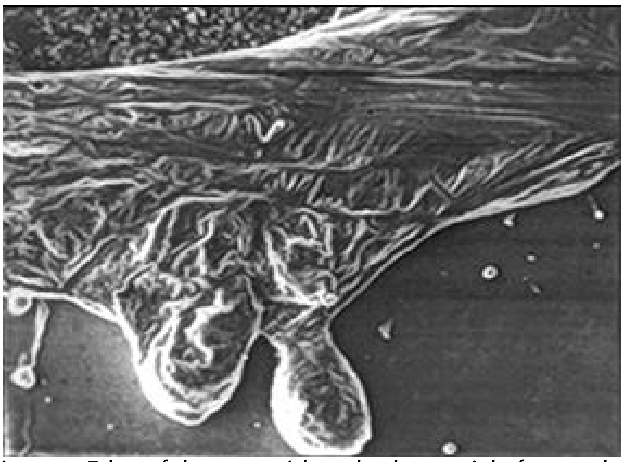


Figure 7. Edge of damage with melted material of amorphe ribon specimen Ti-Co (72%- 88%Co); exposed to ruby laser (λ=0.69μm, τ=30ns, E=0.75J) analyzed by SEM, 257x

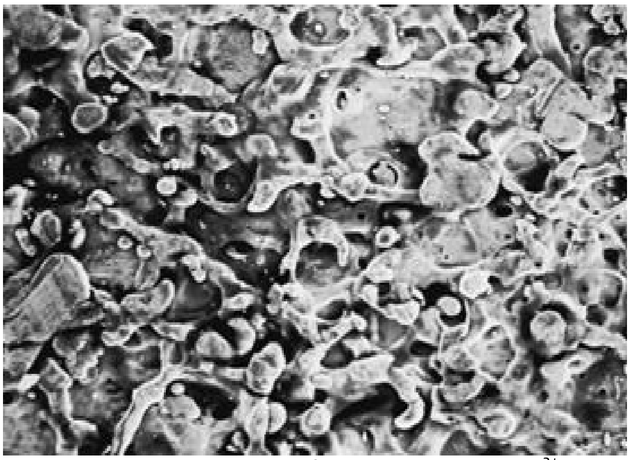
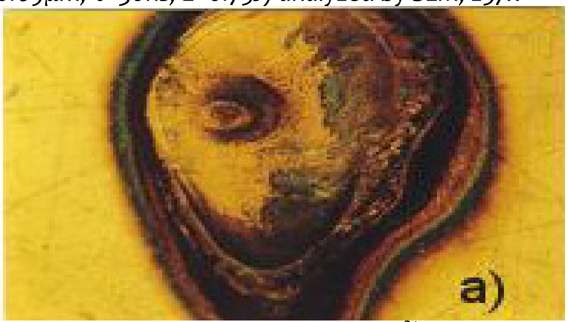


Figure 8. Typical structure of silver exposed to Nd^{3+}:YAG laser in Q-switch regime and some mJ, SEM 1200x



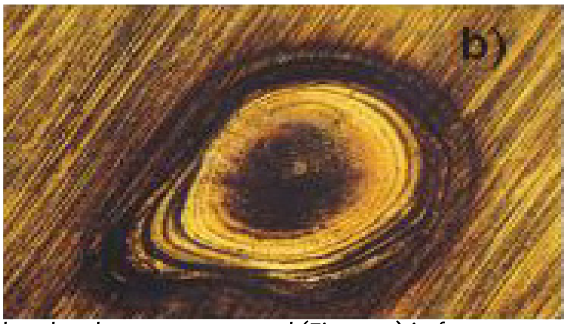
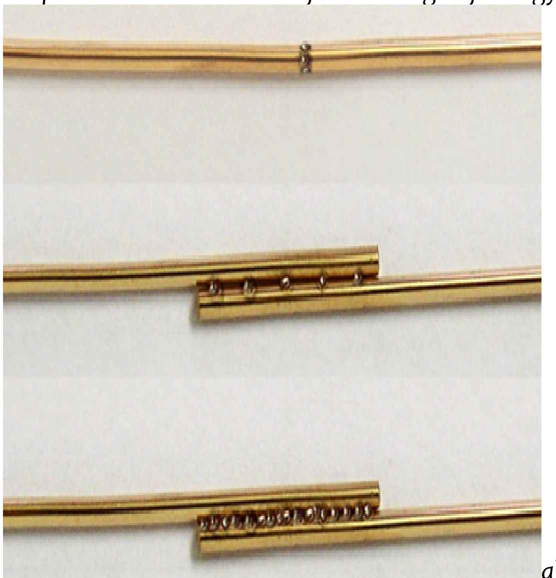


Figure 9a, b. Sample of steel damaged by Nd^{3+}: YAG laser with pulse shape as presented (Figure 4) in free generation regime for unpolished steel substrate for the range of energy of few J. There is interesting colored structure.


a)


b)

Figure 10. Spot laser welding of: a) wires Ø0,5mm and b) strips from gold 585 alloy.(Nd^{3+}:YAG laser)

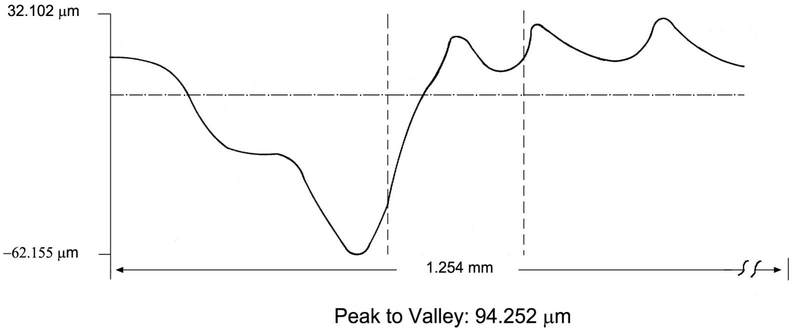


Figure 11. Profyles of the typcal laser crater

The using of laser welding has shown some advantages in many different fields from mining and metallurgy [38], and also in jewel production, when noble metals are used. Two examples of laser welding of noble metals are shown in Figure 10, when one gold alloy (with 58,5mass.%Au) is used.

Laser welding is particaulary available for thiny parts to be joined, as jewel components usualy are. In those circumstances many of the "classical" welding technologies are hardly usable. In Figure 11 some profyle of the typcal laser crater is shown for the damages as in Figure 9.

DISCUSSION AND CONCLUSION

Applicability of certain approximation is essentially limited for the parameters of materials and laser working conditions. Generally, there are two considerations of heat transfer: with and without change of state. More specific calculations do not depend so much on the fact noticed above as on geometry and environmental conditions, etc.

Cu, Al and steel are and were (in the past) the most treated materials in the laser and conventional processing and theoretically studied. Here, pretty good agreement between simple model and experiments for some of them is found and steel is found to be an intermediate case in this scale. These results we present only as demonstration of given approach to most known materials for definite pulse shape obtained without expensive software tools. A complete penetration of the pulse of given characteristics is predicted but the experiments does not confirm the prediction. A complet mechanism of heat losses beyond the scope of the model could be a possible explanation of the fact. Another possible explanation is that the segmentation of the pulses is not fine enough.

The simulation with program Matlab 7.5 with simplified considerations for Dirac δ-pulse gives very fast the first answers about heating processes in materials. A better agreement for interactions with lower power densities is obtained as expected, because the conditions are closer to equilibrium. In many experiments the nature of wrinkles is not whole explained, if it is linked with free generation regime and speckles or not. (The heating and cooling processes depending on laser repetition frequency are present also. Wrinkles structure is obtained in many different laser working regimes.) In our investigations we had many unwanted damage shapes, and with this paper we wanted to show only the results for definite given working conditions and to remain for the approach were phase transformations could be included further after the melting process.

The methods of laser material processing are commonly used in industry. However, the final results of an operation are not easy to achieve. To obtain the range of some laser operation a lot of experimental work is necessary. New materials: compounds, alloys, solid solutions, coating and thin films are, certainly, the great challenge in the cases of very fast processes, and high intensities heat model can be used only if the thermal constants are correctly treated (only electronic parts of conductivity), with including of nonlinearity behavior of constants or the models have to be replaced with hydrodynamics and etc. models.

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