MATHEMATICA MONTISNIGRI
Vol XXIV (2012)

X International Seminar
"Mathematical Models & Modeling in
Laser-Plasma Processes &
Advanced Science Technologies"
LPpM3-2012

LASER-INDUCED PHASE TRANSITIONS IN NEAR-CRITICAL AREA OF ALUMINUM AND MERCURY

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Keywords: Laser-induced phase transitions, critical point, laser ablation.

Annotation. The results of experimental study of phase transitions of aluminum and mercury in the near-critical area of its phase diagram are presented here. Thermodynamical states in the vicinity of critical point of aluminum (theoretical estimations: T_c = 7963 K, P_c = 0.35 GPa) and mercury (T_c = 1763 K, P_c = 0.15 GPa) were obtained using pulsed laser impact of nanosecond duration on metal surface, mechanically confined by a transparent dielectric. This experimental technique gives an ability to obtain pressure pulses with amplitude up to 20 GPa and duration about 10 ns with a tabletop laser setup (with pulse energy up to 1 J).

Q-switched pulsed Nd:YAG laser was used (wavelength 1 μm). During the laser impact synchronous measurements of temperature (optical pyrometer), pressure (piezotransducer) and reflectivity of the metal target were conducted with nanosecond temporal resolution.

The thickness of the irradiated targets varied from several millimeters to tenth fraction of a micron. This parameter considerably influences the character of processes which take place during pulsed laser impact on metal surface. The obtained results show practical possibility of producing near-critical states of aluminum and study of its parameters (temperature, pressure, optical properties).

1 INTRODUCTION

The study of thermophysical, electrical and optical properties of metals in a wide range of applied pressures and temperatures is an important fundamental task that is also significant in many applications such as laser-based material processing, estimation of material resistance during shock-wave impacts, development of new construction materials.

At the current moment there is almost unexplored area for the thermodynamical states of matter with temperatures above 3000 K and pressures above 200 MPa (2000 atm). This region corresponds to the estimated parameters of the critical points of metals, which are the internal scale of the substance. Today, the critical points are experimentally determined only for mercury and alkali metals, reliable experimental data on the parameters of the critical points of the other metals are absent. Also, there are no data on the behavior of metals in the near-critical region: status of the curve of phase equilibrium (binodal) and of the absolute thermodynamic instability (spinodal), the transport properties of metals. The thermodynamic, electrical and optical properties are of great interest – due to relatively high critical temperature, comparable with the potential of ionization, metal vapor at the branches of the binodal can be thermally ionized, so the high-temperature evaporation, apparently corresponds to the transition to the non-ideal plasma state. In the near-critical region phase transitions of the type "metal-insulator transition" is possible.

The most developed static methods for determination of thermodynamic, transport, electrical and optical properties of materials are of limited applicability due to the existence of thermal strength limits of construction materials. Due to this fact dynamic methods are widely spread for the study of states of matter with high temperatures and pressures, including those based on intense laser irradiation. However, the nature of dynamic methods is essentially transient and is usually accompanied by the process of heterogeneous energy input into the system. This feature leads to considerable difficulties in determining the thermodynamic parameters of the system, which continuously changes in time and space.

The absorption of pulsed laser radiation of nanosecond duration leads to heating of the surface layer of the metal and to increase of pressure due to the thermal expansion. However, in the case of a free surface - located in a vacuum or gaseous medium, the efficiency of the process of pressure generation is low – the process of surface unloading takes place. In this paper we propose to use mechanically loaded metal surface - covered with a layer of transparent dielectric with the magnitude of acoustical impedance ρc (ρ - density, c - the speed of sound), comparable to the magnitude of the acoustical impedance of the metal. This considerably increases the efficiency of pressure - more than on two orders of magnitude. This makes it possible to obtain near-critical states of the metals with a table-top laser facility - with a pulse energy of 1 J at a pulse duration of ~ 10 ns[1-2]. An additional advantage of usage of a mechanically loaded metal surfaces is the possibility of collection of thermal radiation in order to determine the surface temperature.

2 LASER HEATING OF METAL AT THE CONFINED SURFACE

At the process of absorption and thermalization of laser radiation in the skin-layer of metal the heat diffusion depth is the following: $\delta_T = \sqrt{\chi \tau_L}$ [3] (for lead δ_T ~ 500 nm, τ_L ~ 10^{-8} s). This value is more than an order of magnitude greater of the laser radiation absorption depth (skin-layer, δ_L ~ 50 nm). So the heating process can be considered as a surface heating with good accuracy. Also the heat diffusion depth at the transverse direction of the heating zone δ_T ~ 500 nm is inessential in comparison with the size of this zone ~ 1 mm and greater (diameter of the laser spot at the metal surface). So the problem can be treated as one-dimensional.

For the absence of phase transitions the temporal dependence of surface temperature of the metal will be determined by the solution of the heat diffusion equation with border conditions of the given heat flux at the surface[4-5]. For the case of laser heating the solution can be written in the following form:

$$P(t) = P_0 + \frac{I_0(1-R)\beta^* c_0}{(1+N)c_p} f(t), \qquad (2)$$

where $\beta^* = \beta\left(1 - 4\frac{c_T^2}{c_0^2}\right)$, β - thermal expansion coefficient, c_0, c_T - transverse and longitudinal sound velocities in metal, N - the ratio of the acoustical impedances of absorbing and transparent media. From (2) it is evident that for the case of constant physical properties of metal the pressure at the metal surface resembles the temporal form of the laser pulse. The acoustical pulse – the pressure wave, which propagates into the depth of the medium will have the same temporal form. That makes it possible to determine the pressure at the sample surface using the acoustical signal which is registered at its rear side.

3 EXPERIMENTAL SETUP

The scheme of the experimental setup is presented at Fig. 1. As a source of laser radiation solid-state Nd:YAG laser (1) working in a Q-switched regime with the following parameters of radiation: wavelength 1.06 um, pulse duration ~ 10 ns, maximum pulse energy ~ 1 J was used. The laser radiation was attenuated by a series of neutral filters (4) after that it was focused by a collecting lenses (f=30 cm) (5) onto the target surface (10) into a spot of 1-3 mm in diameter. The change of the attenuation coefficient and of the distance between the lenses and the target surface allowed to vary, the incident laser radiation energy density in a wide range of values. The uniform distribution of intensity in the laser spot was achieved by a homogenizer (7) (polished glass plates) positioned at the half distance between the lenses and the target. The temporal form of the incident laser pulse was registered with a silicon PIN-diode (6) (rise-time ~ 1 ns). Portion of radiation was directed onto a glass plate (8). The registration of the temporal form of the reflected radiation was made with the analogue photo-diode (11).

The system of registration of thermal radiation consisted of collecting lenses, optical fiber (12), filter from a BGG – glass (14) and photo-transducer (16). The thermal radiation was focused using lenses (9) onto the end of the fiber glass (12). The radiation outgoing from the fiber was collimated by lenses (13) and a BGG-glass filter (14) was put in a parallel beam. Using it the laser radiation of the wavelength of 1.06 um were cut after that the radiation was focused by lenses (15) on a photo-transducer of thermal radiation (16). So the photodetector (16) registered the intensive spectrum of thermal radiation.

The measurement of the acoustical signal (shock wave) from the sample was conducted with a wide-band acoustical piezo-transducer from lithium niobate (10). The electrical signal from the transducer was registered by a digital saving oscilloscope Tektronix TDS 3034. The signals from the photodiodes were registered by another four-channel oscilloscope TDS 3034. The synchronization of both of oscilloscopes was conducted using trigger photodiode (2).

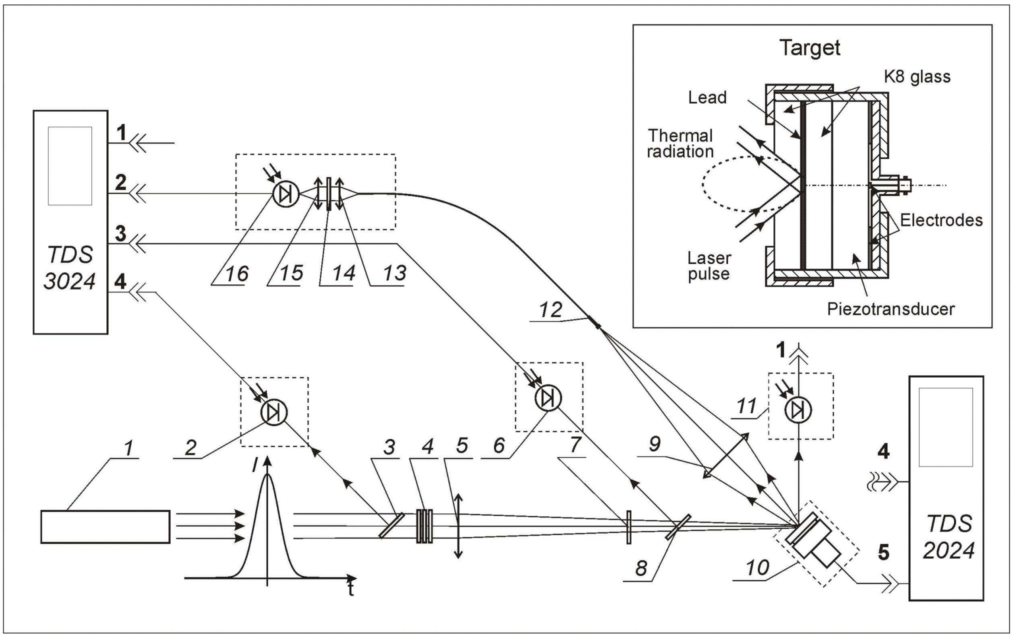

Figure 1: Scheme of the experimental setup

6 EXPERIMENTAL RESULTS

Two reference metals – mercury (Hg) and lead (Pb) were chosen for the studies. The measurements were conducted in a wide range of laser radiation energy density (from 1 mJ/cm^2 to 500 mJ/cm^2, about 40 regimes).

For mercury at Fig. 3a) the normalized profiles of pressure pulses are presented. At Fig. 3 b the absolute values of pressure in semi-logarithmical scale are shown. For lead the same data is presented at Fig. 4 a-b.

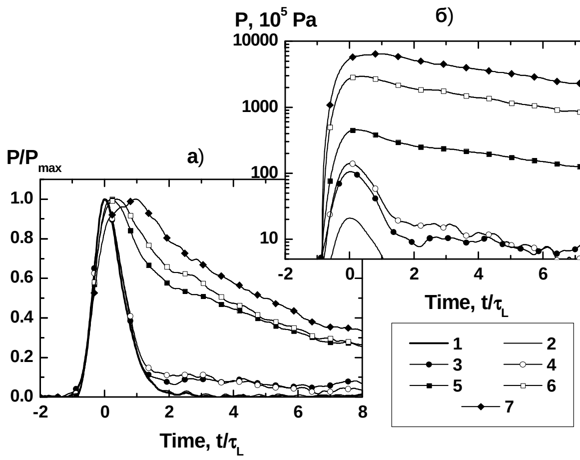

Figure 2: Temporal dependencies of pressure for mercury a) – pressure is normalized at its maximum value, b) – absolute values of pressure. 1 – laser pulse, 2 – E = 6 mJ/cm^2, 3 – E = 32 mJ/cm^2, 4 – E = 38 mJ/cm^2, 5 – E = 83 mJ/cm^2, 6 – E = 200 mJ/cm^2, 7 – E = 270 mJ/cm^2

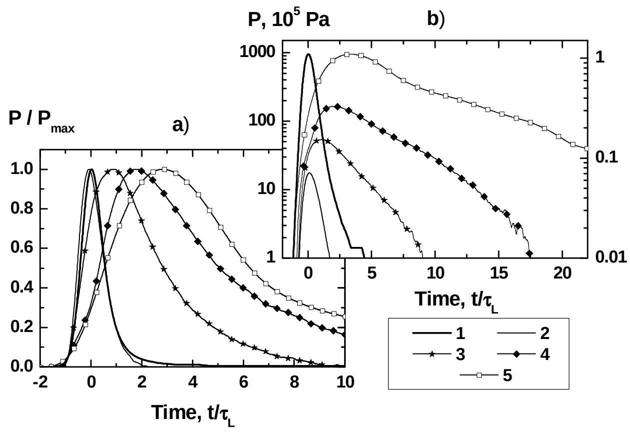

Figure 3: Temporal dependencies of pressure for lead a) – pressure is normalized at its maximum value, b) – absolute values of pressure. 1 – laser pulse, 2 – E = 2.4 mJ/cm^2, 3 – E = 10 mJ/cm^2, 4 – E = 24 mJ/cm^2, 5 – E = 230 mJ/cm^2

In linear (thermoelastical) regimes for low values of laser pulse energy density (up to ~ 4 mJ/cm^2 for lead and ~ 12 mJ/cm^2 for mercury) the temporal form of pressure resembles the shape of the laser pulse according to eq. (2). For greater values of laser radiation energy density the pulse widening (in comparison to the laser pulse) is observed. In this range of thermodynamic parameters of the heated metal the mechanism of pressure generation changes. The "defect of volume" due to phase transition at the heated area and to the appearance of a new phase with less density begins to considerably influence the process of pressure generation. From the definite moment which is explicitly coupled to the threshold value of absorbed laser pulse energy the divergence of the front edges of laser and pressure pulses takes place. This moment determines the beginning of the phase transition. After that the current value of pressure becomes proportional to the value of absorbed energy.

The estimations of correlation between the absorbed energy and phase transition latent heat allow to suppose that in this case the boiling of mercury and the melting of lead takes place. The assumption of boiling of mercury is proved by the relatively high value of pressure (Fig. 2 b, curves 5-7) after the end of the laser impact that can be explained by a slow (~ 90 ns) condensation of vapor. At relatively high values of absorbed energy in lead the rapid phase transition from the heating of the solid phase through melting to boiling takes place (the amount of energy needed for melting is considerably lower). At Fig.3 a, curves 3,4 – the divergence of the leading edges (of laser and pressure pulses), hypothetically due to boiling, begins virtually from the beginning of irradiation. For exact determination of this phase transition as boiling, the range of density change must be known. Besides the general regularity is observed – with the increase of the absorbed energy, besides the widening, the pressure maximums are shifted into area of greater times (Fig. 3 a, lead).

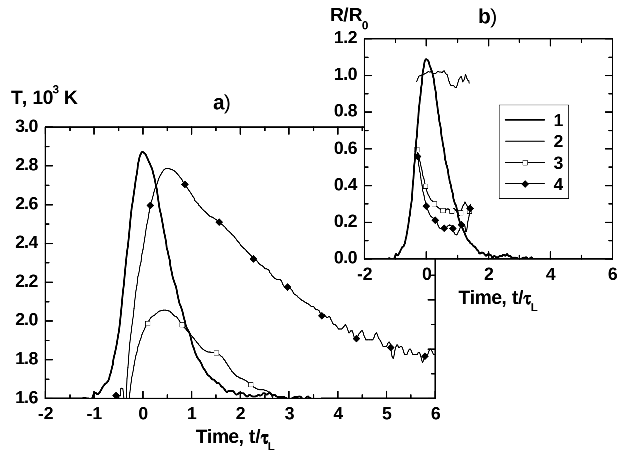

Figure 4: Temporal dependencies of temperature (a) and reflectivity (b) for mercury: 1 – laser pulse, 2 – E = 6 mJ/cm^2, 3 – E = 200 mJ/cm^2, 4 – E = 270 mJ/cm^2

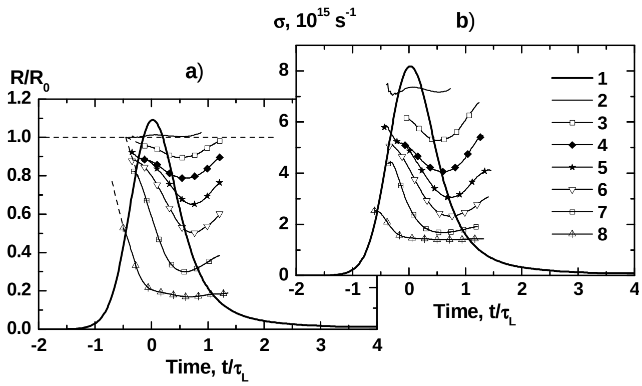

Figure 5: Dynamics of the change of reflectivity (a) and electrical conductivity (b) for lead : 1- laser pulse, 2 – E = 1 mJ/cm^2, 3 – E = 2.4 mJ/cm^2, 4 – E = 5 mJ/cm^2, 5 – E = 7 mJ/cm^2, 6 – E = 10 mJ/cm^2, 7 – E = 24 mJ/cm^2, 8 – E = 230 mJ/cm^2

Simultaneously with pressure measurements the temperature measurements were conducted (only for mercury). The results are presented at Fig. 4. Using the data on dynamics of temperature and pressure change at the mercury surface it is possible to plot the diagram of the laser heating process in P-T coordinates (Fig. 6). At this figure curves 4,5 are plotted using experimental data. It is clear, that in this regimes for considerably low values of absorbed energy the supercritical states of mercury were obtained with high values of temperature (T ~ 2400 K) and pressures (P ~ 0.7 GPa). Linear regimes (curves 1-4) which had no experimental data on temperature are obtained using eq. (1)-(2).

The dynamics of the relative change of reflectivity at the laser irradiation wavelength at the time of laser impact with the angle of incidence of 45 degrees for mercury and lead in relative units (R/R_0) are presented at Fig. 4 b and Fig. 5 correspondingly. The initial value of reflectivity coefficient for mercury was R_0 = 0.50 and for lead R_0 = 0.60. With an increase of incident laser irradiation power density the reflectivity (at time scale comparable with the duration of the laser pulse) decreases more than five times (Fig. 5) This considerable decrease of reflectivity and correspondingly the increase of absorbed energy during the duration of the laser pulse τ_L demands accounting of this effect in all calculations during the processing of experimental data. At Fig. 5 b the estimation of temporal dependence of conductivity obtained using data on reflectivity change and the Drude model is presented. In the limits of this model the order of the conductivity decrease is the same as the decrease of reflectivity. The considerable decrease both of the conductivity and reflectivity can be explained by the substantial density decrease for high levels of absorbed energy that takes place with an increase of temperature in a thin subsurface layer of metal.

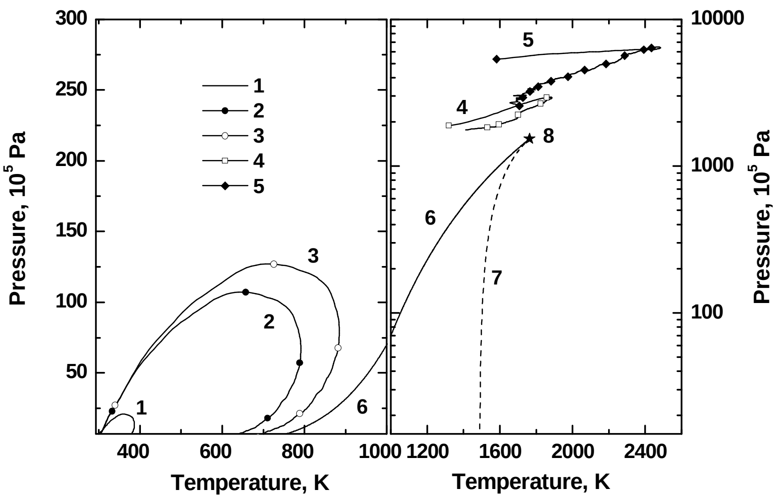

Figure 6: P-T diagram of laser heating of mercury: 1 – E = 6 mJ/cm^2, 2 – E = 32 mJ/cm^2, 3 – E = 38 mJ/cm^2, 4 – E = 200 mJ/cm^2, 5 – E = 270 mJ/cm^2, 6 – binodal, 7 – liquid spinodal, 8 – critical point

After experiments on the test metals (mercury and lead) the developed method was applied to the study of aluminum in the near-critical area of its phase diagram. Surface of aluminum targets of different thickness (from several mm to the tenths of microns) was covered by a layer of transparent dielectric (natrium silicate). The results are presented at the figures 7-10.

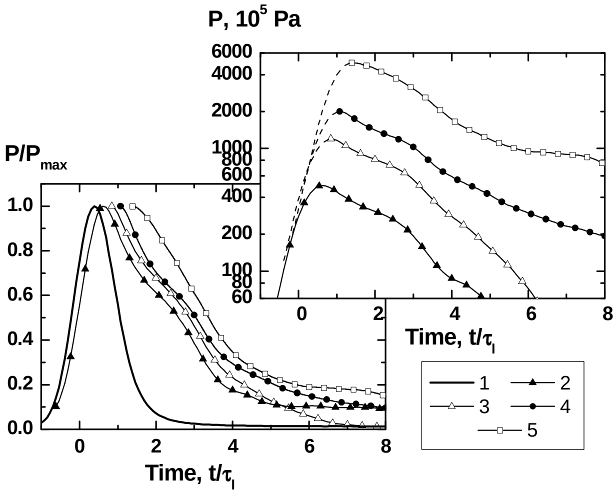

Figure 7: Temporal dependencies of normalized (a) and absolute (b) pressure in aluminum: 1 – laser pulse, 2 – E = 1 J/cm^2, 3 – E = 2 J/cm^2, 4 – E = 3 J/cm^2, 5 – E = 5 J/cm^2

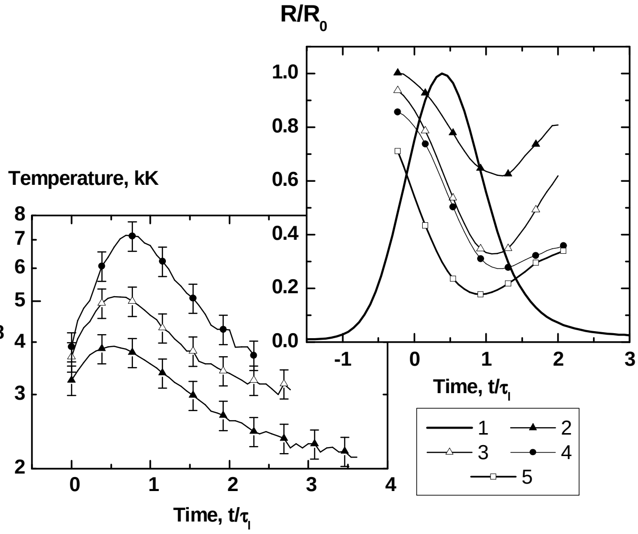

Figure 8: Temperature and reflectivity of the aluminum surface: 1 – laser pulse, 2 – E = 1 J/cm^2, 3 – E = 2 J/cm^2, 4 – E = 3 J/cm^2, 5 – E = 5 J/cm^2

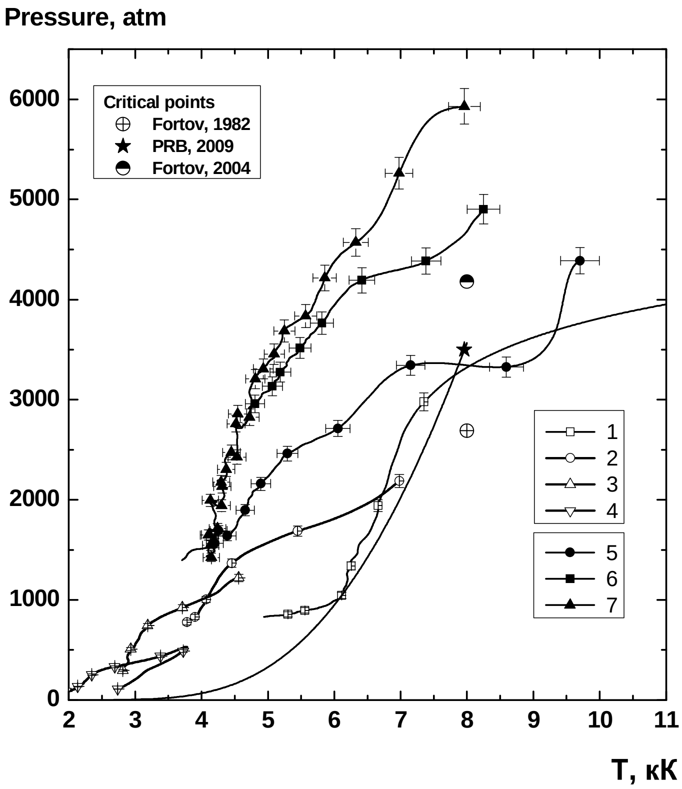

Figure 9: Process of the laser heating of aluminum in P-T coordinates: 1 - E = 5 J/cm^2 (N = 6), 2 - E = 3 J/cm^2 (N = 6), 3 - E = 2 J/cm^2 (N = 6), 4 - E = 1 J/cm^2 (N = 6), 5 - E = 5 J/cm^2 (N = 1.6), 6 - E = 5 J/cm^2 (N = 1.3), 7 - E = 5 J/cm^2 (N = 1)

7 CONCLUTIONS

The presented results show the efficiency of using optoacoustical method in conditions of confined geometry for inducing high-energy states of metals and studying phase transitions.

It is shown that before the start of melting the pressure is proportional to the intensity of the laser radiation and after crossing the melting threshold it becomes proportional to the value of absorbed laser radiation energy. The moment of deformation of the leading edge of pressure pulse determines the start of melting.

It is obtained that in the studied range of laser irradiation energies with the beginning of phase transition the electrical conductivity of the heated metal decreases considerably that leads to decrease of the reflectivity of the thin sub-surface layer of metal more than 5 times.

It should be noted that the suggested method can be successfully applied in three important areas of study:

- for inducing and studying of metastable and unstable states in the sub-critical area of the phase diagram;

- for intense shock waves generation to study the shock-plastic deformation of metals;

- for the study of optical properties in a wide range of temperatures (~ 1 eV) and densities (~ 1 g/cm^3). In this range the absorption properties of materials are almost unknown.

LITERATURE

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