Physics of Extreme States of Matter — 2014

Moscow, 2014
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This compendium is devoted to investigations in the fields of thermal physics of extreme states of matter and physics of high energy densities. Different models and results of theoretical calculations of equations of state for materials at high pressures and temperatures, physics of shock, detonation and combustion waves, interaction of intense laser, x-ray and microwave radiation, powerful particle beams with matter, techniques of intense energy fluxes generation, experimental methods of diagnostics of ultrafast processes, low-temperature plasma physics, issues of physics and power engineering, as well as technology projects are considered. The majority of the works has been presented at the XXIX International Conference on Equations of State for Matter (March 1–6, 2014, Elbrus, Kabardino-Balkaria, Russia). The edition is intended for specialists in physical and technical problems of power engineering.

This conference is dedicated to the centenary of birth of academician Yakov Borisovich Zel’dovich (March 8, 1914–December 2, 1987).

The conference is sponsored by the Russian Academy of Sciences and the Russian Foundation for Basic Research (grant No. 13-02-06212).

The editorial board announces with deep regret the death of the colleagues and friends, Dr. Vladimir Vladimirovich Milyavskiy (July 22, 1969–June 12, 2013), who was the organizing and program committees member of the Conferences on Equations of State for Matter and Interaction of Intense Energy Fluxes with Matter, and Prof. Gennady Vasil’evich Sin’ko (February 7, 1950–September 8, 2013), who was a regular and active participant of these conferences starting with one of the first meeting.

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POWER INTERACTION WITH MATTER

FROZEN NANOSTRUCTURES PRODUCED BY ULTRASHORT LASER PULSE
Khokhlov V. A.1,*, Inagamov N. A.1, Anisimov S. I.1, Zhakhovsky V. V.2, Emirov Yu. N.3, Ashitkov S. I.2, Komarov P. S.2, Agranat M. B.2

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Surface nanostructures, arising under the influence of a short laser pulse on a metal target as a result of nucleation, stretching and then freezing of foam-like material in the surface melt, are considered.

Keywords: Short pulse laser ablation, surface nanostructures, frozen nanofoam

Introduction. When we speak of the surface structures arising under laser irradiation, the first thing that usually comes to mind is a regular ripple-like structure arising from the interaction of the incident and surface electromagnetic waves. The properties of such structures are defined by the resonance condition between the incident and surface waves. Further, such structures will not be considered.

We will study irregular structure, the cause of which, as a rule, is the development of thermal and hydrodynamic inhomogeneities. A typical representative of such structures is a foam-like structures remaining on the surface of the target after a short laser pulse.[1]

Experimental results [1]. In the described experiments the study of deformation of the target surface in the area, heating by femtosecond (100 fs) pulse (FLP) of Cr:forsterite laser system, was performed using femtosecond interference microscopy.[2] There is a crater on the target surface after exposure in the case if laser fluence in spot center $F_c$ exceeds ablation threshold $F_{abl}$. Fig. 1 shows an example of the crater interferogram at $F_c > F_{abl}$. The structure of the surface layer of the target with a residual deformation was investigated by Transmission Electron Microscopy (TEM) by preparing a thin membrane (shown schematically in Fig. 1, which is then analyzed by a transmission microscope Technai TF 20. As a result there the existence of nanocavities observed inside deformed surface layer, Fig. 2

2T stage. Fast heating of electrons by short laser pulse converts the metal in two-temperature (2T) state with the electron temperature $T_e$ is much greater than the ion (phonon) temperature $T_i$. An underlying concept of 2T state of metal with hot electrons and cold ions was suggested in the paper [3]. To solve 2T heat conduction equations the accurate values of an electron-ion coupling parameter $\alpha$ [4-6] and electron heat conduction $\kappa, \chi = \kappa/c_e$ [5-7] are required, here $c_e$ is electron heat capacity. The required data have been obtained from solutions of kinetic equations in r-approximation [6, 7]. Reliable description of the 2T conduction is necessary since $\kappa_{2T} \sim 10^4$ ~ $10^5$ ~ $10^6$ $\text{W} \cdot \text{K}^{-1} \cdot \text{m}^{-1}$ is very high (far beyond $\kappa_{1T} \sim 100$ $\text{W} \cdot \text{K}^{-1} \cdot \text{m}^{-1}$) and $\kappa$ together with $\alpha$ defines thermal depth $d_T \sim \sqrt{\chi T_{eq}} \sim 100$ nm. Thus $d_T$ turns out few times larger than an optical skin depth $\delta \approx 10-20$ nm.[8] Here $T_{eq} \sim c_i/\alpha \sim 2-10$ ps is an electron-ion temperature equilibration time, ion heat capacity $c_i \approx 3k_B n_{atom}$, $\alpha \sim 10^{17}$ $\text{W}^{-1} \cdot \text{m}^{-3}$ [4, 6], $d_T \sim \nu_F \sqrt{T_{eq} \tau_e}, \tau_e = 1/\nu_e$, where $\nu_F$ is Fermi velocity, $\nu_e = \nu_{ei} + \nu_{ee}$ is electron collision frequency.

Within a 2T stage, which lasts $t_{eq}$, the speed of heat propagation (thermal wave) is very large $v_{th} \sim c_e (\nu_F/c_e) / \sqrt{\tau_e / t}$ because Fermi velocity $v_F$ and its Mach number are large $Ma_F = v_F/c_e \sim 100$. For flu-
Figure 3. Spatial distributions of temperatures of ion sub-system $T_i$, electron thermal flux $q_e$, taken with inverse sign, and power of electron-ion energy transfer $W_{ei}$ per unit of volume. Al film was irradiated from the left side through a glass substrate. Initially glass was at $x < 0$, Al film was placed at $0 < x < 1.2$ μm. Laser pulse has duration $\tau_L = 100$ fs, time is reckoned from a maximum of a pulse, energy $E_{abs} = 0.13$ J/cm$^2$ is absorbed in a skin layer of $\delta = 15$ nm. Arrow marks shock in glass. We neglect light absorption and heat conductivity in glass. Therefore, the ion heating $W_{ei}$ decreases sharply at the glass-Al interface.

Figure 4. Melting and isochoric curves of Al together with profiles at $2T$ ($t = 0.5$ ps) and hydrodynamic ($t = 20$ ps) stages. The end point 3' sits inside a melt. Its position is $x = 95$ nm ($t = 20$ ps). The $T$, $\rho$ values for inner melt and glass ($x < 95$ nm) are not shown for $t = 20$ ps.

too slow at this time scale. This is a reason why the melting temperatures $T_i$ in points "L" and "S" in Fig. 3 are higher than the triple point temperature 0.954 kK for Al. Profiles of thermo-mechanical flow on the phase plane $T$ - $\rho$ are shown in Fig. 4. Within the 2T stage at $t = 0.5$ ps the profile 1-S-L-2 is almost coincide with a curve corresponding to initial density of Al. The point 1 gives a room temperature state of Al far from glass-Al boundary. The points S and L present solidus and liquidus, as in Fig. 3. Maximum of $T_i$ (the point 2 in Fig. 4) is achieved near a glass-Al boundary, see Fig. 3. Dependence $p_i(T_i)$ at $t = 0.5$ps is shown as the profile 1-S-L-2 in Fig. 4. This curve can be presented also in a parametric form: $T_i(x)$, $p_i(x)$. Near a boundary the density of Al drops due to expansion in direction of glass. At this early stage the expansion of Al is driven mainly by electron pressure $p_e$.

Spatial positions of point S and L at pressure $p_i$ and temperature $T_i$ profiles in Figs. 3 and 4 coincide during supersonic propagation of heat. Images of those points on pressure profile in the hydrodynamic "h" stage are marked as $L_i$ and $S_i$ in Fig. 4. The "h" stage follows the 2T stage. At the "h" stage those images are split off from their positions in 2T stage - comp. $L_n$, $S_n$ versus L, S in Fig. 4. The images propagate along acoustic characteristics carrying values of pressure corresponding to isochoric melting. But temperatures in points $L_{ih}$ and $S_{ih}$ decrease during propagation because the points move to the right along the $x$ axis, as shown in Fig. 3, and temperature decreases in this direction. As a results, a gap between the ($L_{ih}$, $S_{ih}$) and (L, S) line segments is formed on the $T$ - $\rho$ plane. We refer to this process as emission of compression wave by decelerating thermal wave.

Let’s call the front "L" marked in Fig. 3 as a melting front, because there is pure liquid to the left side relative to this point. At a moment shown in Fig. 3 the front velocity is 63 km/s! This is hypersonic velocity, therefore, liquid do not have time to decrease its density after melting (isochoric regime), and ion pressure increases during fast heating (expansion release is
sects a two-phase solid-liquid melting zone along differently directed line segments on a thermodynamic phase plane. On a $\rho - T$ plane those segments rotate from isochoric to isobaric directions during transition from 2T to 1T flow. The feature with inverse dependence of $T_i$ in Fig. 5 isn’t connected with mechanical reasons - namely, with a release of shear stress in a melting zone. It also exists in the pure plastic hydrodynamic simulations, see Fig. 5 in [8]. This feature is linked to the finite heat of fusion and expenses of energy on melting. Small deviations from exact local equilibration $T_s = T_i$ remains in 1T stage, see Fig. 5. They are necessary for thermal transport from hot to cold places, because ion heat capacity $c_i$ is larger than electron capacity $c_e$ while heat in metals is carried mainly by electrons (ratio of thermal diffusivities $\chi_i/\chi_e$ is small).

Resolidification and surface nano-structures

Time scales of thermal and hydrodynamic processes taking place in a surface layer of target are: $t_{eq} \sim 2-7$ ps - duration of 2T stage; $t_{shd} \sim 20$ ps - hydrodynamic stage; $t_{sl} -$ stopping of motion near surface; and $t_{sol} \sim 1$ ns - solidification of molten metal. For fluences higher than the nucleation threshold $F > F_{nuc}$ [1] the scale $t_{sol}$ increases sharply from small values $t_{shd}$ to a nanosecond time scale. This is caused by large difference in rigidity between continuous melt and foam-like material. In case with bubbles the near surface velocities drop down to $\sim 10 - 100$ m/s for $F > F_{nuc}$ and $t \sim 1$ ns. Thus, $t_{sol}$ becomes $t_{sol}$ and situations with freezing of slowly moving nanostructures may realize [1]. The corresponding picture of frozen bubbles beneath the surface obtained in molecular dynamics (MD) simulation [9, 10] is shown in Fig. 6.

A result of analysis a wide range of MD calculations for different levels of pumping energy they can be be divided into three qualitatively different segments: (i) $F < F_{nuc}$, (ii) $F_{nuc} < F < F_{abl}$ and (iii) $F_{abl} < F$. 

Figure 5. Layer of molten Al near glass-Al boundary. Electrons transfer energy from a hot place and therefore become colder than ions in this hot place. This picture is interesting since it shows how the 2T case $T_s \gg T_i$ tends to a limit where description through Stefan task becomes valid in metals: $c_e/c_i \ll 1, \kappa_e/\kappa_i \gg 1$.

Figure 6. Expansion and freezing of foamed aluminum. As a result of electron cooling of molten surface layer the resolidification front moves up through continuous melt and reaches the bottoms of bubbles. Due to supercooling of surface liquid at later time, solid nuclei are also formed inside the melt and at free surface. (a) cavitation in liquid surface layer shown on density map; (b-e) atomic order parameter shows phase state of material: green colors correspond to solid state, while red colors correspond to liquid; (c) crystallization begins at liquid-solid interface first, then in bulk of supercooled melt and at free boundary of melt; (d) crystallization of foam walls initiated by formation of solid nuclei on free liquid surfaces of walls; (e) liquid walls are broken, a tiny droplet forms at the end, and liquid jet moves back to the target along partially frozen pike.

Figure 7. Focused ion beam/transmission electron microscopy (FIB/TEM, lamella technique) cut of nickel surface. The lamella has been taken from a lateral part of a crater produced by a single optical laser shot. The grey layer on the top is a protecting layer of Pt organic. Dark knobs are frozen jets. Bright crescent are deformed frozen nanobubbles similar to the bubbles observed in Al target, see Fig. 2.

In case (ii) “frozen foam” can be observed, see Fig. 7. In the paper [1] fluence corresponds to the case (iii), in the central region of the laser spot and the case (ii) on the periphery.

In the case (iii) we call such structures “frozen broken foam”.

Such structures is obtained also with X-ray lasers [11, 12]. In the X-ray case the plasmonic effects are negligible. Therefore, the X-ray structures remain chaotic (no ripples) after any number of laser shots $N$. The performed experiments confirm this.

Optical lasers produce similar structures [13-17]. Our analysis shows that they appear if $F > F_{nuc}$ and $N \sim 1$.

This is a key explaining thermo-mechanical mechanism of appearance of chaotic surface nano-structures. The existence of similar structures and a wide range
of wavelengths of laser from the optical to the X-ray confirms their thermo-mechanical fluctuation nature.

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