Strength of iron melt at high extension rate during femtosecond laser ablation

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Abstract. Time and spatial-resolved interferometric technique in a picosecond range was used for continuous registration of motion of iron target surface heated by femtosecond laser pulse. The magnitude of the tensile stress $0.5–1.3$ GPa leading to fracture of molten iron at the strain rate of $\sim10^9$ s$^{-1}$ was experimentally determined from the measured velocity histories of the spalled layer movement.

1. Introduction

Today, femtosecond laser is an important tool for fundamental investigations of nonequilibrium processes in condensed matter. Our understanding of the interaction of femtosecond laser pulse (FLP) with metals is based on the two-temperature model [1]. Since the heat capacity of the electrons is much smaller than that of the lattice, an ultrashort laser pulse with duration less than the heating time of the lattice can heat electrons to a very high temperature while the lattice remains relatively cool. Isochoric heating of thin surface layer of metal target gives rise to powerful tensile stresses and spalling of a part of surface liquid nanolayer of a material. The nature of the fracture is a cavitation process of a formation and growth of vapor nuclei in a metastable stretched melt [2–7].

In the current work, we experimentally determine the value of the tensile stresses $\sigma_{spl}$ that leads to the fracture of liquid iron after irradiation by FLP in the vicinity of ablation threshold using temporal and spatial resolved interferometric technique. Determination of a value of $\sigma_{spl}$ is performed by measuring the velocity profile of a free surface of a target [8]. For continuous recording of motion in a picosecond range, the frequency domain (chirped) interferometry [9,10] is used. The application of this method allows the continuous registration of process dynamics during single exposure [11].

2. Experiment

The optical scheme of our measurement system is shown in figure 1. A source of femtosecond pulses was a Ti:sapphire laser system generating pulses of 40 fs at wavelength 795 nm. A small part of the amplified chirped pulse was taken from laser beam before the compressor and used for diagnostics. A powerful part of the pulse of femtosecond duration after the compression was applied to heat the target. The experimental scheme was described in detail earlier [10].
Figure 1. Schematic diagram of the measurements: 1—focusing lens; 2—target; 3—Michelson interferometer; 4—spectrometer; 5—CCD camera; 6—energy meter.

Experimental samples were iron film with a thickness of 1 µm deposited on glass substrates by magnetron sputtering.

Pump pulse of p-polarization was focused on the target surface at an angle of incidence of 60° using a lens with a focal length of 30 cm. Spatial distribution of energy density in a focal spot was Gaussian with radius $r_0 = 30 \mu m$ at the level of $e^{-1}$.

For the detection of the hydrodynamic motion of the spalled layer of iron target, we use a frequency modulated (chirped) pulse with a duration of about 300 ps at a central wavelength $\lambda = 795$ nm and spectral width $\Delta \lambda = 40$ nm. The experimental scheme provides a continuous registration of process dynamics with a temporal resolution of $\delta t \approx 2$ ps in the range $0 \leq \Delta t \leq 200$ ps.

The diagnostic part of the setup is a Michelson interferometer, where one of the mirrors was the surface of the iron sample. An objective with NA = 0.3 was used to transfer the image of rear surface to the entry slit of a diffraction spectrometer Acton 2300i. The interferograms after spectrometer were recorded by 12 bit CCD camera SensiCam QE. The interferometer was adjusted in such a way that the interference fringes were perpendicular to the entry slit.

Fourier processing of interferograms allows to obtain an accuracy of measurement of phase changes of the reflected probe wave approximately $\delta \varphi \approx 0.01$ rad and correspondently provides an error of the surface displacement $\delta z \approx 2$ nm. The magnitude of the surface displacement $\Delta z$ associated with the phase changes was determined from the relation:

$$\Delta z = \frac{\lambda \Delta \varphi}{4\pi}. \quad (1)$$

The CCD camera recorded interferograms before, during and after the irradiance of target by FLP. The algorithm of processing of interference patterns is described elsewhere [12].

3. Results and discussion

The value of ablation threshold $F_a$ was obtained from the measured dependence of ablation crater size on the laser pulse energy [13]. Simultaneously, the reflection coefficient $R$ of pump beam was measured. The corresponding values were equal to $F_a \approx 0.087 \pm 0.01$ J/cm$^2$ and $R \approx 0.4$. So the absorbed pump energy for iron at the ablation threshold in this case is equal to $F_a^{abs} = (1 - R) F_a \approx 0.05$ J/cm$^2$. 
Figure 2. Histories of the amplitude (1) and phase (2) changes of the probe pulse at normalized energy density of the pump pulse $F/F_a = 1.2$.

Figure 3. The surface displacement as a function of time at $F/F_a = 1$ (1), 1.1 (2) and 1.2 (3).

Figure 2 represents temporal profiles of amplitude $\Delta A(t)$ and phase $\Delta \varphi(t)$ measured in the central part of the interaction area. Rapid change of amplitude and phase during the first several picoseconds, compared with a time of electron–phonon energy exchange $t_{ei}$, is obviously associated with a heating and melting of the lattice. After this, the hydrodynamics stage begins. Subsequent smooth decrease in reflectance is due to the movement of a thin spalled layer with a thickness less than the skin depth of probe.

Figure 3 shows the temporal dependences of the displacement of the surface $\Delta z(t)$, which describe the dynamics of the expansion of the surface layer of iron target after the exposure by FLP with different energy density.

All profiles in figure 3 were measured in a single shot and plotted for the center (curve 3) and peripheral parts (curves 1 and 2) of the interaction area. Figure 4 represents the velocity profiles of iron target surface movement as functions of time. These profiles are obtained by differentiating of the displacement dependences, which was calculated using relation (1).
Figure 4. Velocity history of surface layer expansion of iron sample at different fluences of FLP: $F/F_a = 1$ (1), 1.1 (2) and 1.2 (3).

The maximum of the velocity $u_{\text{max}} \approx 0.15 \text{ km/s}$ is reached about 10 ps after the beginning of the movement. An intensive acceleration at this range is due to gradient of pressure in the isochorically heated surface layer. Further decrease of the velocity in time interval from 10 to 30 ps is associated with the resistance of a substance to the action of tensile stresses arising during its motion.

In the case of thermomechanical ablation, the temperature of a heated surface layer achieves the value of $\sim 2$–3 kK, and a homogeneous melting occurs at time scale of $\sim 10^{-12}$–$10^{-11}$ s [14]. If the arising tensile stress exceeds the magnitude of strength of the melt $\sigma_{\text{spl}}$, the spalling occurs in the molten layer of the material, which results in fracture of the melt during its stretching. After the formation of the spalled layer, it moves mechanically with approximately constant speed, the magnitude of which depends on the excess of FLP energy density over the ablation threshold and increases with increasing $F$. As one can see in figure 3, near $F_a$, the inertial velocity of spalled layer is close to zero and increases to about 0.1 km/s at the energy density of FLP $F/F_a \approx 1.2$.

A value of the tensile stress $\sigma_{\text{spl}}$ that causes the fracture of the material under extension can be approximately evaluated from linear acoustic expressions [8]:

$$\sigma_{\text{spl}} = \frac{1}{2} \rho_{\text{liq}} c_{\text{liq}} \Delta u. \quad (2)$$

Here, $\Delta u$ is the pullback velocity (see figure 4), $\rho_{\text{liq}}$ and $c_{\text{liq}}$ are the density and the speed of sound in the melted metal respectively. Figure 5 represents the measured dependence of $\Delta u$ from the energy density of FLP.

The value of ablation threshold in metals and semiconductors approximately 1.5–2 times exceeds the melting threshold (melting temperature of iron is $T_m = 1809$ K). Let us roughly estimate the temperature $T_{\text{liq}}$ of molten layer of the iron target in vicinity of ablation threshold from the absorbed energy according the expression

$$T_{\text{liq}} \approx \frac{(1 - R) F d_T^{-1} - \Delta H_m}{\rho_0 c_p} + T_0, \quad (3)$$

where $d_T$ is the molten layer thickness; $\rho_0 = 7.87 \text{ g/cm}^3$ is the initial density; $c_p = 0.46 \text{ J/(g K)}$ is the specific heat capacity; $\Delta H_m = 13.8 \text{ kJ/mol}$ is the melting heat; $T_0 = 300$ K. We assume
that the thickness $d_T$ of iron melt is twice greater than the depth (of 25 nm) of a crater, which is finally formed on the irradiated surface of the sample. So, $d_T \approx 50$ nm. This value is in a good agreement with the result of molecular-dynamic simulation for iron [7]. Results of the temperature estimation by (3) are listed in table 1.

The density and the speed of sound of the liquid phase of iron at the estimated temperatures were calculated with use of equation-of-state model [15]. Then, the spall strength values were obtained from relation (2). The strain rate of melt under spalling is evaluated as follows [8]:

$$
\dot{\varepsilon} = \frac{\Delta u}{\Delta t} \frac{1}{2c_{\text{liq}}},
$$

(4)

Here $\Delta t = t_{\text{max}} - t_{\text{min}}$, $t_{\text{max}}$ is the time corresponding to the maximum value of the velocity in figure 4, and $t_{\text{min}}$ is a moment corresponding to the first minimum, $\Delta t = 19$ ps in the case. All the evaluations are presented in table 1.

Thus, we obtained that, at increasing the relative fluence $F/F_a$ from 1 to 1.2, the value of tensile strength $\sigma_{\text{spl}}$ decreases from 1.3 GPa to 0.5 GPa as well as the strain rate decreases from 0.9 to 0.4 ns$^{-1}$.

A thickness of a spalled layer near the ablation threshold is evaluated from the dynamics of expansion (figure 4) using the relation

$$
L_{\text{spl}} = \frac{1}{2} c_{\text{liq}} \Delta t.
$$

(5)

So, the thickness of spalled layer is $L_{\text{spl}} \approx 40$ nm. This value corresponds well with the measured crater depth of 25 nm.

Table 1. Results of measurements and estimations.

| $F/F_a$ | $\Delta u$, km/s | $T_{\text{liq}}$, kK | $\rho_{\text{liq}}$, g/cm$^3$ | $c_{\text{liq}}$, km/s | $\sigma_{\text{spl}}$, GPa | $\dot{\varepsilon}$, $10^8$ s$^{-1}$ |
|--------|------------------|----------------------|-----------------------------|-------------------------|--------------------------|-------------------------|
| 1      | 0.12             | 2.9                  | 6.53                        | 3.40                    | 1.3                      | 9.3                     |
| 1.2    | 0.045            | 3.2                  | 6.36                        | 3.37                    | 0.5                      | 3.5                     |

Figure 5. Dependence of the pullback velocity from normalized fluence of FLP for iron.
4. Conclusion
The dynamic tensile strength of iron melt has been evaluated from the velocity history of spalled layer measured in a picosecond range using time resolved interferometry. The obtained results in vicinity of ablation threshold indicate the decreasing of strength with the increase of temperature of the melt. The obtained dynamic tensile strength of iron melt 0.5–1.3 GPa at the strain rates $(4–9) \times 10^8 \text{s}^{-1}$ and temperature about 3 kK are in good agreement with results of simulations [7].

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