Measurements of strength of metals in a picosecond time range

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Abstract. We studied the shock-wave phenomena in metal films of a micron or submicron thickness irradiated by femtosecond laser pulses. The single-shot interferometer technique was used to record the time and spatial resolved displacements of rear surfaces of the films. The free surface displacement histories were converted into the free surface velocity histories using several various approaches. As a result, new data on the shear and spall strength have been obtained for aluminum (3.2 GPa and 8.2 GPa) and iron (7.9 GPa and 20.3 GPa) in strongly metastable states close to their ultimate values.

1. Introduction
With the advent of high-power femtosecond lasers it has become possible to study the material behavior at extremely high strain rate. The first such studies on aluminum [1-4] revealed the possibility of generation and measurements of shear and tensile stresses close to their ultimate possible (“ideal”) values [5-7]. It is also important, that the experimental data sets obtained by different methods over a wide range of the loading rate are described by the unified dependence of the deformation and fracture on the strain rate [8]. Unlike fcc aluminum, iron has more rigid bcc crystal structure. At a pressure of 13 GPa iron undergoes a polymorphic $\alpha \rightarrow \varepsilon$ transition. The high-pressure $\varepsilon$-phase has a hexagonal close-packed structure. Molecular dynamics simulations [9] indicate the coincidence of the onset of plastic deformation and polymorphic $\alpha \rightarrow \varepsilon$ transformation. The corresponding values of stress are sensitive to the used potential of interatomic interaction and varied with the direction of shock compression.

In this work we continue our previous investigation of laser driven high-rate deformations in metals in a picosecond time range using chirped pulse interferometry [10]. Unlike multipulse pump-probe method [2] this single shot technique provides a much higher reliability of the measurements.

2. Experiment
The investigated samples were polycrystalline 99.99% aluminum (500, 760, 1200 nm thick) and iron (250, 540 nm thick) films deposited by magnetron sputtering on 150-μm borosilicate glass substrates. The film thickness was measured with an atomic force microscope with an accuracy of ±10 nm.

A Ti:sapphire femtosecond laser system was used for generation and diagnosing of the shock wave phenomena. After the regenerative amplifier a chirped pulse of the system was split into two parts. A
weaker frequency modulated probe pulse of 300 ps duration at the central wavelength $\lambda_0 = 795$ nm with bandwidth FWHM $\Delta \lambda = 23$ nm was used for monitoring the sample free surface motion. More powerful part was compressed to a pulse of 100 fs duration with total energy up to 2 mJ at wavelength 795 nm. The shock generating pump pulse was focused through the glass substrate onto the surface of the metallic film. The spatial distribution of the pump laser radiation in the focal spot was Gaussian with a diameter of around 40 $\mu$m at the $e^{-1}$ level.

The diagnostic part of the setup was a Michelson interferometer, where one of the mirrors was the rear surface of the sample. An objective with NA = 0.2 was used to transfer the image of rear surface to the input slit of the Acton 2300i diffraction spectrometer. The interferometer was adjusted in such a way that the interference fringes were perpendicular to the input slit. The slit image was positioned on the sample surface through the centre of the breakout region. The interference pattern at the output of the spectrometer was recorded by a CCD-camera. The system was calibrated in time using a varied optical delay line. The wavelength in a recorded spectrum was juxtaposed to time, whereas the other axis (along the slit) of the interferogram corresponded to spatial coordinates on a target. The values of temporal and spatial resolution were 1 ps and 2 $\mu$m respectively.

In each experiment a CCD camera recorded three interferograms: before, during, and after the shock wave breakout. 2D Fourier processing of interferograms [11], include a procedure of normalization of the initial-transient phase distributions, provides accuracy of a phase shift measurement on a level of about $\approx 0.01$ rad, which corresponds to 1–2 nm of the error in the surface displacement.

3. Results of measurements

Figures 1 and 2 present the free rear surface displacement histories $z(t)$ which have been measured for iron samples of 250 nm and 540 nm thickness at the laser fluence in the central part of the focal spot $F_0 = 3$ J/cm$^2$. Figure 2 summarizes the displacement histories obtained from the central area of the breakout profile for several laser shots. For all figures zero time moment was chosen arbitrarily. The results demonstrate high reproducibility of the measurements in the initial stage of the surface motion. A small relative shift of the profiles in time is probably due to small variation of the sample thickness.

A significant discrepancy of the histories at delays more than 50–80 ps with respect to the beginning of motion is associated with the process of spall fracture, which will be discussed below.

![Figure 1](image1.png)  
**Figure 1.** Phase shift data of shocked iron samples of 250 nm and 540 nm in thicknesses.

![Figure 2](image2.png)  
**Figure 2.** The arrays of the free surface displacement profiles of shocked iron samples of 250 nm and 540 nm, measured in a central part of an interaction area.
After the initial stage of order 10–20 ps we observe further increasing of surface motion speed that indicates two-wave configuration of shock.

The free surface velocity profiles $u_{fs}(t)$ were evaluated by differentiating the measured $z(t)$ dependence with the subsequent iteration procedure, as a result of which the integral $u_{fs}(t)$ is the best fit of the measured displacement history (figure 3).

Figure 3. The example of processing of the displacement profile for aluminum samples of 760 nm in thickness measured in the centre of the focal spot at incident laser fluence 2.1 J/cm$^2$; dash lines show the measured $z(t)$ profile and the free surface velocity history obtained by differentiating the smoothed $z(t)$ profile. The solid lines show the results of the iteration procedure.

Figure 4 shows free surface velocity histories averaged over four shots for iron (a) and aluminum (b) films of different thicknesses at driven laser pulse fluence $F_0 = 3$ J/cm$^2$. Results of the measurements for iron clearly demonstrate a split of the shock wave into a two-wave configuration and its strong decay on submicron propagation distance. The speed of the first shock wave, determined from the measured time interval between the shock arrivals, has been found equal to $U_S = 6.45 \pm 0.2$ km/s in the 250–540 nm section.

The surface velocity in first shock wave decreases from $1.06 \pm 0.06$ km/s at a distance of 250 nm to $0.45 \pm 0.03$ km/s at 540 nm. A comparison of the shock wave speed and particle velocity $u_p = u_{fs}/2$ shows that the measured value $U_S$ is much higher than calculated using known Hugoniot. Taking into account the small ($\leq 1$ ps) rise time of the first wave, we can certainty conclude, that the registered two-wave configuration consist of elastic precursor and plastic shock waves. Using the $U_S$ and $u_p$ values averaged out of several shots the metastable elastic Hugoniot of iron has been determined as $U_S = 5.97 \pm 1.2 u_p$ km/s. After that, the Hugoniot elastic limit $\sigma_{HEL} = \rho_0 U_S u_p$ was found to be $27.5 \pm 2.5$ GPa and $11.0 \pm 1$ GPa at the propagation distance of 250 nm and 540 nm respectively. Here $\rho_0$ is the material density.
For aluminum samples the reliable evidence of a two-wave structure under these conditions was not found. The shock wave speed was found equal to $U_S = 7.1 \pm 0.3 \text{ km/s}$ in the 500–1200 nm section. In the case of aluminum we recorded high propagation speed and did not record the splitting of the shock wave. Under described conditions, the detected shock wave was purely elastic. Similarly, the metastable Hugoniot of elastic compression was estimated as $U_S = 6.44 + 1.4 \nu_p$, that well coincides with our previous data [2, 5]. The value of the shock stress in this case decreased from 14.3 GPa at 500 nm of the propagation distance down to 7.7 GPa at 1200 nm.

Figure 5 presents the results of measurement on the stress-strain plane. The value of deviatoric stress $\sigma - p$ reached 4.3 GPa for aluminum and 10.5 GPa for iron. Corresponding values of shear stress at uniaxial compression $\tau = \frac{3}{4}(\sigma - p)$ [12, 13] were estimated as 3.2 GPa and 7.9 GPa for Al and Fe respectively.

![Figure 5](image)

**Figure 5.** Recorded states in elastic shock waves (points) in comparison with Hugoniot of iron (a) and aluminum (b).

Figure 6 shows the sets of velocity histories for iron and aluminum of various thicknesses measured for one shot. The waveforms are plotted for the different distances from the center of the focal spot and correspond to incident laser fluence varied from 2.5 J/cm$^2$ to 1 J/cm$^2$.

![Figure 6](image)

**Figure 6.** Evolution of shock in iron (a) and aluminum (b) for incident laser fluence varied from 2.5 J/cm$^2$ to 1 J/cm$^2$.

The smooth growth in the upper part of the compression shock wave after sharp rise is not necessarily the result of the onset of plastic deformation and can be associated with the intermediate stage of the shock wave formation for the given energy density of driving laser pulse.
The spall strength value $\sigma_{\text{spall}}$ was evaluated using measured velocity pullback (figures 3, 4 and 6) assuming purely elastic or elastic-plastic behavior of the material [2]. Estimations have been done accounting for nonlinear compressibility of the material. For 250 nm thick iron and 500 nm thick aluminum samples in the center of the focal spot plastic deformation contributed to the waveforms and the $\sigma_{\text{spall}}$ was estimated with correction on distortion of the waveforms in elastic-plastic materials [14]. Estimations of the spall layer thickness of ~100 nm and ~200 nm indicate that the spallation probably occurred at the boundary of the molten layer. The maximal values of strength for aluminum were registered for the sample of 760 nm in thickness (figure 3).

Figure 7 shows the measured values of spall strength $\sigma_{\text{spall}}$ of iron and aluminum as a function of the strain rate in a comparison with the earlier experimental and calculated data (see [2, 15-18] and the references).

![Figure 7](image_url)

**Figure 7.** Measured spall strength of iron (a) and aluminium (b) in comparison with the previously obtained data; full and open circles represent strength in a “cold” and “hot” states respectively.

4. Discussion

The results, obtained for aluminum in this work with the single shot technique are in good agreement with the previous data, obtained using a multi-pulse interferometric microscope [1]. This relation between shock and particle velocities indicates elastic shock compression of aluminum up to 14 GPa and iron up to 27.5 GPa in a picosecond time domain. Estimated values of shear stress of 3.2 GPa for aluminum and 7.9 GPa for iron are close or even exceed the ideal strength values for these materials. Observed values of the spall strength of aluminum reached 7.7 GPa at a strain rate of order $(2\sim3)\times10^9$ s$^{-1}$ and 20.3 GPa for iron.

The ultimate value of shear resistance – “ideal” shear strength obtained through *ab initio* calculations for iron is 7.2–7.5 GPa [19, 20]. It should be noted that the compression leads to increase of the shear modulus and the value of “ideal” shear strength. The tensile strength of iron from the first-principle calculations [20, 21] is about 27.7–28.45 GPa, which is slightly greater than the maximal value $\sigma_{\text{spall}} = 20.3$ GPa, according to the given measurements. Under uniaxial tensile deformation, the “ideal” tensile strength must be somewhat lower than at volume stretch [21]. Moreover, *ab initio* calculations of ideal shear and bulk strength have been done for a temperature of 0 K, while it is known that the increase in temperature reduces these values.

Thus in this work, the stressed states of aluminum and iron, very close to the values of the “ideal” strength were measured and implemented in the picosecond range of load duration. Unfortunately, the non-stationary shock and the high speed of the relaxation processes do not allow estimating the final state of iron after compression in two waves. For this reason, the possibility of polymorphic phase transition in the picosecond range of compression remains open and requires further investigation.
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References
[1] Ashitkov S I, Agranat M B, Kanel G I and Fortov V E 2010 JETP Lett. 92 516
[2] Whitley V H, McGrane S D, Eakins D E, Bolme C A, Moore D S and Bingert J F 2011 Appl. Phys. 109 013505
[3] Crowhurst J C, Armstrong M R, Knight K B, Zaug J M and Behymer J F 2011 Phys. Rev. Lett. 107 144302
[4] Ashitkov S I, Agranat M B, Kanel G I and Fortov V E 2012 AIP Conf. Proc. 1426 1081
[5] Sin’ko G V and Smirnov N A 2002 JETP Lett. 75 184
[6] Jahnátek M, Hafner J and Krajičí M 2009 Phys. Rev. B 79 224103
[7] Clatterbuck D M, Krenn C R, Cohen M L, Morris J W 2003 Phys. Rev. Lett. 91 135501
[8] Kanel G I 2012 AIP Conf. Proc. 1426 939
[9] Kadau K, Germain T C, Lomdahl P S and Holian B L 2005 Phys. Rev. B 72 064120
[10] Ashitkov S I, Komarov P S, Ovchinnikov A V, Struleva E V and Agranat M B 2013 Quantum Electronics 43 242
[11] Temnov V, Sokolovskii-Tinten K, Zhou P and Linde D 2006 J. Opt. Soc. Am. B 23 1954
[12] Zeldovich Ya B and Raizer Yu P 1967 Physics of Shock Waves and High Temperature Hydrodynamic Phenomena (New York: Academic)
[13] Kanel G I, Razorenov S V and Fortov V E 2004 Shock-Wave Phenomena and the Properties of Condensed Matter (New York: Springer)
[14] Kanel G I 1999 Fatigue Frac. Eng. M. 22 1011
[15] Kanel G I, Razorenov S V and Fortov V E 2005 Mech. Solids 40 69
[16] Zhilyaev P A, Kuksin A Yu and Stegailov V V 2010 Phys. Solid State 52 1619
[17] Zhakhovsky V V and Inogamov N A 2010 JETP Lett. 92 521
[18] Ashitkov S I, Komarov P S, Agranat M B, Kanel G I and Fortov V E 2013 JETP Lett. 98 384
[19] Clatterbuck D M, Chrzan D C and Morris J W 2003 Acta Materialia 51 2271
[20] Ogata S, Li J, Hiroasaki N, Shibutani Y and Yip S 2004 Phys. Rev. B 70 104104
[21] Cerny M and Pokluda J 2007 Phys. Rev. B 76 024115