PAPER • OPEN ACCESS

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To cite this article: P S Komarov et al 2019 J. Phys.: Conf. Ser. 1147 012023

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Generation of giant elastic ultrashort shock waves in chromium films by femtosecond laser pulses

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Abstract. Using the ultrafast chirped interferometry of picosecond resolution, the evolution of superelastic ultrashort shock waves has been studied in chromium films over propagation distances ranging from 440 to 1160 nm driving by laser pulses of 450 fs duration. The relationship between the shock wave and the particle velocity indicates the shock compression is elastic up to 23 GPa. The experimental data on a metastable elastic Hugoniot in this range were obtained. The achieved shear stress behind the shock wave 8.1 GPa is close enough to the calculated “ideal” shear strength of chromium.

1. Introduction
In the last decade a significant attention has been paid to investigation of a high rate deformation and fracture of materials under extremely short shock loading which can be realized with laser pulses of pico- and femtosecond duration [1–9]. This interest, on the one hand, is due to fundamental questions of mechanics and physics of condensed matter, and on the other hand, it is associated with the development of high precision laser processing and nanostructuring of materials. It is known [10] that tensile and shear strength of materials increases with increasing of a deformation rate. There are exist maximum permissible values of the “ideal” strength of the substance [11], the closest to which in the experiment can be approached by increasing the strain rate. In experiments with ultrashort laser pulses, extremely high rates of deformation at the level of $10^9–10^{10}$ s$^{-1}$ are implemented. The application of ultrashort laser pulses makes it possible to investigate the elastic-plastic properties of metals at extremely high values of shear stresses comparable with the shear modulus. Chromium is a high-strength body centered cubic (bcc) metal with the bulk modulus of 300 GPa and the shear modulus of 110 GPa. The data obtained in this paper allowed us to extend for the first time the elastic Hugoniot of chromium in the region of metastable elastic states to compression stress of about 23 GPa.

2. Experiment
In the article, to study the evolution of ultrashort shock waves in chromium, an interferometric technique using a subnanosecond frequency-modulated (chirped) diagnostic pulse was applied to investigate the dynamics of deformation of the target surface in the picosecond range in a single shot mode with spatial and temporal resolution [12, 13]. The investigated samples were polycrystalline chromium (440, 810, 1160 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 $\pm 10$ nm. A Ti:sapphire femtosecond laser system was used for generation and diagnosing of the shock wave phenomena. Generation of compression pulses was carried out by heating the frontal surface of the sample with laser pulses through a transparent glass substrate. Diagnostics of shock wave phenomena was carried out on a free back surface. In this configuration, the magnitude of the created stress is limited by the optical breakdown into substrate. Therefore, in order to obtain higher compression, the duration of laser pulses was increased up to 450 fs by adjusting the compressor. A frequency-modulated chirped pulse was split into two parts. A weaker frequency modulated probe pulse of 300 ps duration at the central wavelength of $\lambda_0 = 795$ nm with bandwidth $\Delta \lambda = 23$ nm was used for monitoring the sample free surface motion. The shock driven powerful pump pulse with total energy up to 5 mJ was focused through the glass substrate onto a metal–glass interface (figure 1). The spatial distribution of the pump laser radiation in the focal spot was Gaussian with a diameter of about 40 $\mu$m at the $e^{-1}$ level.

The diagnostic part of the setup was a Michelson interferometer, where as one of the mirrors, the rear surface of the sample served. An objective with NA = 0.3 was used to transfer the image of the rear surface to the input slit of imaging diffraction spectrometer (Acton 2300i) equipped with an 600 g/mm grating. 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 (charge-coupled device) 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 interference pattern 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 interference patterns: before, during, and after the shock wave breakout. Two-dimensional Fourier processing of interference patterns, 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 an error of the surface displacement.
Figure 2. Phase shift data of shocked Cr sample of 810 nm in thicknesses irradiated by pulses of 450 fs duration with the fluences of $F_0 = 3.0$ J/cm$^2$. Colorbars show phase shift in radians.

Figure 3. The same as in figure 2 but for $F_0 = 4.2$ J/cm$^2$.

3. Results
Figures 2 and 3 represents time and spatial resolved phase distributions measured at the free rear surface of the chromium film sample of 810 nm in thickness after shock wave arrival, driven by pulses of 450 fs duration with a spatial Gaussian distribution and various fluence at the center of the focal spot $F_0$. For this configuration of the experiment, there is a limitation on the intensity
Figure 4. A measured breakout profile at a free surface after shock arrival for \( F_0 = 3.0 \text{ J/cm}^2 \) at time delay 0, 6.6, 11, 27, 47 and 69 ps.

Figure 5. A measured breakout profile at a free surface after shock arrival for \( F_0 = 4.2 \text{ J/cm}^2 \) at time delay 0, 6.6, 11, 27, 47 and 69 ps.

of the incident laser radiation in connection with the possible occurrence of nonlinear effects and optical breakdown in glass, which can be seen in figure 3 at a fluence of \( F_0 = 4.2 \text{ J/cm}^2 \).

The corresponding breakout profiles for various time delay and different fluence are shown in figures 4 and 5.
Figure 6. The arrays of the free surface displacement profiles of shocked chromium samples of 440 (1), 810 (2) and 1160 nm (3) at laser fluence $F_0 = 3.0 \text{ J/cm}^2$.

Figure 6 summarizes the displacement histories $z(t)$ obtained from the central area of the breakout profile for several laser shots (thin grey lines). The thick colored lines are the results of averaging. 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 (about 1 ps) is probably due to small variation of the sample thickness. The free surface velocity profiles $u_{fs}(t)$ were evaluated by differentiating the measured $z(t)$ dependencies 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 7).

Figure 8 shows the free surface velocity histories for the samples of different thickness, which were obtained after the processing of the averaged dependencies $z(t)$ plotted in figure 6.

A shock wave is of triangle shape with a sharp leading edge. The maximum free-surface velocity decreases from 0.87 km/s at 440 nm to 0.48 km/s at 1160 nm, and the width slightly increases from 17 to 22 ps. The smaller duration of generated compression pulse in Cr in compare to Al [1,13] at a similar conditions may be explained by a lower value of heat conductivity [14].

4. Discussion

Figure 9 represents elastic and plastic Hugoniot curves for Cr. Here the points are obtained from the experiment. The particle velocity $u_p$ corresponds to the half of the maximum free surface velocity value. The obtained parameters of the shock wave deviate significantly from the plastic Hugoniot $U_{s}^{pl} = c_b + bu_p$, where $c_b = 4.8 \text{ km/s}$—the bulk sound velocity; $b = 1.55$ [15]. The most probable reason for this is that in the experiments performed in the picosecond range of loading durations, plastic deformations do not develop, and the detected compression wave is elastic. This assumption confirms an extremely short rise time of the shock wave. Using the experimental data and the value $c_l = 6.64 \text{ km/s}$ for the longitudinal speed sound in chromium [16], the Hugoniot for the elastic compression can be represented in the form $U_{s}^{el} = c_l + bu_p$, where coefficient $b = 1.4$. 
Figure 7. The example of processing of the displacement history of chromium sample of 440 nm in thick.

Figure 8. Evolution of the ultrashort shock wave in chromium at the propagation distances of 440 (1), 810 (2) and 1160 nm (3).

The elastic stress at uniaxial compression $\sigma_z$ can be determined from the expression:

$$\sigma_z = \rho_0 U_s \epsilon_1 u_p,$$

(1)

where $\rho_0 = 7.19$ g/cm$^3$ is the material density. The corresponding value of $\sigma_z$ is equal 23 ± 1; 16 ± 1 and 12.0 ± 1 GPa at a propagation distance of 440, 810 and 1160 nm respectively. The
obtained results are in a good agreement with the data of elastic precursor decay in chromium from [16]. The achieved maximum shear stress \( \tau \) is determined from the difference between the longitudinal elastic compression stress \( \sigma_z \) and pressure \( p \) at a given specific volume \( V \) according to the relation [17]:

\[
\tau = \frac{3}{4}(\sigma_z(V) - P(V)).
\]

The measured maximal value \( \tau = 8.1 \) GPa is close enough to calculated value of the “ideal” shear strength of chromium equal to 13.4 GPa.

5. Conclusion
The evolution of superelastic shock waves with duration of about 20 ps generated by femtosecond laser pulses in the micron and submicron chromium film samples was investigated using ultrafast chirped interferometry. It is found that uniaxial shock compression is elastic up to stress of 23 GPa or more in this range. The shear stresses behind the shock wave reach 8.1 GPa, which is approximately 60% to the calculated value of the “ideal” shear strength of chromium.

Acknowledgments
The experiments were carried out on the facility “Terawatt Femtosecond Laser Complex” JIHT RAS. This work was supported by the Russian Science Foundation, grant No. 14-50-00124.

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