Dynamic response of molybdenum to ultrafast laser induced shock loading

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Abstract. Molybdenum is a bcc transitional metal whose high-pressure behavior has attracted considerable experimental and theoretical interest. In the present paper, a chirped pulse interferometry was applied to study shock waves evolution in molybdenum under ultrafast loading created by a femtosecond laser. The elastic compression up to 17 GPa on a submicron propagation distance was achieved. Also the spall strength of molybdenum film samples was estimated to be 30–40% of ultimate tensile strength value at the strain rate of $10^9$ s$^{-1}$.

1. Introduction
In recent years the behavior of materials under mechanical loads of very short duration, generated by pico- and femtosecond lasers, has received increased attention [1–10]. Such studies are of interest for forecasting intense pulsed actions on materials and constructions in a wide range of load parameters, and for the development of wide-range equations of state of matter. It is known that the flow stress and fracture resistance of solids increases with increasing loading rate. The present work is a continuation of the study of the behavior of metals under action of loads in a picosecond range generated by the femtosecond laser. In particular, the use of such loads allows most closely approach the theoretical limit of shear and tensile strength.

Molybdenum (Mo) is a refractory high-strength structural material, widely used in industry. Investigation of its temperature-strength properties in the sub-microsecond range was dedicated a number of works [11–16]. Molybdenum is one of the most high-strength metals (elastic and shear modulus of molybdenum are 330 and 120 GPa, respectively) and is quite complex object to study its resistance to deformation and fracture.

2. Experiment
The samples under investigation were polycrystalline molybdenum films of 95% purity deposited by magnetron sputtering on 150-µm borosilicate glass substrates. Molybdenum films (420, 750, 1080 nm thick) had texture-oriented structure with the size of grains on the surface of order 20–50 nm. 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 of loading. For the diagnosing of the shock wave phenomena chirped pulse interferometry was applied. The shock generating
Figure 1. Spatial-time distribution of phase at free rear surface of Mo film sample of 750 nm in thickness during shock arrival; laser fluence $F_0 \approx 0.6$ J/cm$^2$.

pump pulse with the duration of 160 fs was focused through the glass substrate onto the surface of the Mo film in a spot with Gaussian distribution and diameter of 30 µm at the $e^{-1}$ level.

The implemented diagnostics ensured the continuous recording of a displacement of a free rear surface of a sample as a function of time in the range $t \leq 200$ ps with the lateral spatial resolution $\delta y \approx 3$ µm and the temporal resolution $\delta t \approx 1$ ps. The two-dimensional Fourier processing of interference patterns, including a procedure of normalization of the initial-transient phase distributions, provides an accuracy of the displacement $\delta z \approx 1–2$ nm. The technique of measurements described in detail elsewhere [17].

3. Results of measurements

Figure 1 illustrates the reconstructed spatial-temporal distribution at free rear surface of Mo sample 750 nm in thick after shock wave arrival, generated by laser pulse with the value of fluence in the central part of the focal spot $F_0 \approx 0.6$ J/cm$^2$. Corresponding peak intensity of laser radiation $I \approx 4 \times 10^{12}$ W/cm$^2$ is less that a damage threshold of the glass substrate.

Figure 2 summarizes histories of the displacement $z(t)$ obtained at the central area of the breakout profile for several laser shots. The results demonstrate high reproducibility of the measurements in the initial stage of the surface motion. A large discrepancy of the histories at delays more than 20–30 ps with respect to the beginning of motion is associated with the process of spall fracture.

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

Figure 4 presents the evolution of compression wave at a propagation distance from 420 to 1080 nm. The zero time corresponds to the beginning of motion of the rear surface of film sample 420 nm in thick.
Figure 2. The array of $z(t)$ profiles measured at the free rear surface of shocked Mo sample.

Figure 3. The displacement (gray line) and free surface velocity (blue line) profiles for Mo sample of 750 nm in thickness measured in the centre of the focal spot at an incident laser fluence 0.6 J/cm$^2$. 
Figure 4. Velocity histories $u_{fs}(t)$ at the free rear surface of molybdenum film samples with the different thicknesses: 420 (1), 750 (2) and 1080 nm (3).

4. Discussion

The profiles $u_{fs}(t)$ had triangular shape with the small rise time of order 2 ps. The measured amplitude $u_{fs}(t)$ decreased from 0.64 to 0.36 km/s, and width (FWHM) increased from 25 to 55 ps. The absence of the wave splitting into elastic-plastic two-wave configuration point out that plastic deformations were not developed and the detected shock wave is purely elastic. This assumption is supported by the small rise time, which indicates an insignificant contribution of dissipative processes.

On the other hand the detected shock wave velocity is unexpected low. The averaged velocity of the shock wave, determined from the measured time interval between the shock arrivals, has been found equal to $U_s = 5.3 \pm 0.2$ km/s as in the 420–750 nm and in the 750–1080 nm sections. This value is essentially less, than the longitudinal speed of sound was found to be $c_l = 6.46$ km/s for bulk samples [18]. Perhaps such a discrepancy may be explained by texture-oriented structure of the tested molybdenum film samples.

The peak shock stress, determined from the expression $\sigma = \rho_0 U_s u_{fs}/2$ (where $\rho_0 = 10.2$ g/cm$^3$ is the normal density of Mo) decreased from 17.3 GPa at 420 nm of the propagation distance down to 13.0 GPa at 750 nm and to 9.7 GPa at 1080 nm.

The dynamic tensile strength of the material just before spall fracture, i.e., the so-called spall strength $\sigma_{spall}$ in an acoustic approximation can be obtained from the recorded velocity history (see figure 4) using the expression: $\sigma_{spall} = \rho_0 U_s \Delta u_{fs}/2$. Here the pull-back velocity $\Delta u_{fs}$ is the difference between the peak and minimum value of the free surface velocity (see figure 3). The determined pull-back velocities in the present experiments with the molybdenum samples of 420, 750, 1080 nm in thick were 0.47, 0.43 and 0.29 km/s (see figure 4). The corresponding spall strengths are equal to 12.7, 11.6 and 7.8 GPa at strain rate $\dot{\varepsilon} \sim (0.3-1) \times 10^9$ s$^{-1}$. Here, the strain rate is defined as a rate of the material expansion: $\dot{\varepsilon} = \Delta u_{fs}/(2c)$. 
In accordance with the expression \( L_{\text{spall}} = c(t_{\text{min}} - t_{\text{max}})/2 \), a plane spall fracture in the experiment with a sample of 420 nm is located at a distance of \( \approx 90 \) nm from the sample rear surface (the same as for samples 750 and 1080 nm \( L_{\text{spall}} \) is equal to 190 and 240 nm, respectively). The estimations of \( L_{\text{spall}} \) show that under considered conditions fracture takes place in a solid state.

The observed increase of the dynamic tensile strength is fitted in figure 5 by the dependence: \( \sigma_{\text{spall}} = 0.18 \dot{\varepsilon}^{0.2} \).

The scanning-electron-microscope images of cross-sections of the samples showed a columnar structure (with the columns oriented normally to a substrate and dimensions 20–50 nm across). In case of short load duration fracture may occur locally at the grain boundaries and inhomogeneities of structure and not develop to the complete destruction similar a spallation plate. To complete a destruction additional energy on the growth of discontinuities and plastic deformation of the material around them is required. It may be more reasonably occurs in hard materials as molybdenum. In particular, it may be the reason of the observed variation of the dependences \( z(t) \) and weak spall pulses at the velocity profiles.

5. Conclusions

With the use of chirped pulse interferometry, the evolution of shock compression wave generated by femtosecond laser pulses in polycrystalline Mo film samples was investigated at a submicron propagation distance. The absence of wave splitting and small rise time show that compression is elastic up to 17.3 GPa at a distance of 420 nm. The obtained in this work spall strength of molybdenum film samples is about 30–40\% from ultimate bulk strength, which value according to the results of ab initio calculations [19–21] is in the range 28.8–43.2 GPa.
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References
[1] Ashitkov S I, Agranat M B, Kanel G I, Komarov P S and Fortov V E 2010 JETP Lett. 92 516–20
[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 E M 2011 Phys. Rev. Lett. 107 144302
[4] Ashitkov S I, Komarov P S, Agranat M B, Kanel G I and Fortov V E 2013 JETP Lett. 98 384–8
[5] Demaske B J, Zhakhovsky V V, Inogamov N A and Oleynik I I 2013 Phys. Rev. B 87 054109
[6] Crowhurst J C, Reed B W, Armstrong M R, Radonsky H B, Carter J A, Swift D C, Zaug J M, Minich R W, Teslich N E and Kumar M 2014 J. Appl. Phys. 115 113506
[7] Ashitkov S I, Komarov P S, Struleva E V, Agranat M B and Kanel G I 2015 JETP Lett. 101 276–81
[8] Abrosimov S A, Baizhulin A P, Voronov V V, Geras’kin A A, Krasyuk I K, Pashinin P P, Semenov A Yu, Stuchebryukhov I A, Khishchenko K V and Fortov V E 2013 Quantum Electron. 43 246–51
[9] Inogamov N A et al 2013 Contrib. Plasma Phys. 53 796–810
[10] Krasyuk I K, Pashinin P P, Semenov A Yu, Khishchenko K V and Fortov V E 2016 Laser Phys. 26 094001
[11] Hixson R S and Fritz J N 1992 J. Appl. Phys. 71 1721–8
[12] Duffy T S and Ahrens T J 1994 J. Appl. Phys. 76 835–42
[13] Furnish M D and Chhabildas L C 1992 The minerals, metals, and materials society High Strain Rate Behavior of Refractory Metals and Alloys ed Asfahani R et al (Warrendale: Academic Press) p 229
[14] Chhabildas L C, Barker L M, Asay J R and Trucano T G 1990 Int. J. Impact Eng. 10 107–24
[15] Senchenko V N, Belikov R S and Popov V S 2015 J. Phys.: Conf. Ser. 653 012100
[16] Senchenko V N, Belikov R S and Popov V S 2016 J. Phys.: Conf. Ser. 774 012020
[17] Ashitkov S I, Komarov P S, Ovchinnikov A V, Struleva E V, Zhakhovskii V V, Inogamov N A and Agranat M B 2014 Quantum Electron. 44 535–9
[18] Zaretsky E B and Kanel G I 2016 J. Appl. Phys. 120 105901
[19] Luo W, Roundy D, Cohen M L and Morris J W J 2002 Phys. Rev. B 66 094110
[20] Ogata S, Li J, Hiroasaki N, Shibutani Y and Yip S 2004 Phys. Rev. B 70 104104
[21] Krenn C R, Roundy D, Morris Jr J W and Cohen M L 2001 Mater. Sci. Eng., A 319 111–4