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LARGE PROFILED MOLYBDENUM SINGLE CRYSTALS PRODUCED BY ADDITIVE PLASMA-INDUCTION TECHNOLOGY AS A PROMISING MATERIAL FOR HIGH-POWER LASER OPTICS COMPONENTS

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This work presents the results of a study on large profiled molybdenum single crystals produced by additive plasma-induction technology, aimed at their application as materials for high-power laser optics components. The choice of molybdenum as a promising material for mirrors and thermally loaded optical elements is justified by its high melting point, low coefficient of linear expansion, high thermal conductivity, and strong reflectivity in the infrared range. It is shown that conventional melting and shaping techniques lead to polycrystalline structures, which limit the achievable mirror surface quality. The proposed additive plasma-induction technology enables the growth of large-size molybdenum single crystals with controlled crystallographic orientation and geometry, suitable for manufacturing large-area mirrors. A comparative study of the optical properties of samples with (110), (111), (100), and (112) planes was carried out by analyzing laser diffuse scattering diagrams (λ = 633 nm). It was found that the mirror surface quality strongly depends on the crystallographic orientation: the smallest diffuse scattering angle ($2\Theta < 10°$) is observed for the (110) plane, which is the most suitable for forming mirror elements of high-power laser optics. The obtained results are consistent with literature data for small single crystals and confirm the feasibility of using large molybdenum single crystals produced by plasma-induction technology in high-power laser systems.

INTRODUCTION

The development and operation of modern high-power laser systems are accompanied by stringent requirements for optical components – mirrors, reflectors, and heat-loaded elements. These components must not only ensure efficient interaction with laser radiation over a wide spectral range (1...10 μm) but also maintain structural and optical stability as well as beam resistance under intense thermal and mechanical stresses.

Among the materials suitable for manufacturing such components, molybdenum stands out due to its unique combination of a high melting point, low coefficient of linear thermal expansion, high thermal conductivity, and significant reflectivity in the infrared range. Molybdenum mirrors exhibit excellent resistance to most aggressive environments – from exposure to seawater and high humidity to the effects of chemical agents.

It is also important that molybdenum demonstrates high resistance to sputtering under the action of charged particle beams, such as ion bombardment, which makes it attractive for manufacturing mirrors used to control laser beams in Thomson scattering diagnostics in plasma devices such as the LHD [1, 2]. These properties make molybdenum a promising material for the fabrication of mirror-type heat-loaded elements in high-power laser systems (Table 1).

Table 1
Main physico-chemical properties of molybdenum affecting its optical characteristics

Parameter	Value	Reference
Melting point	2623	[3]
Density	10.28	[3]
Thermal conductivity at 20 °C	~138	[4]
Electrical conductivity, μΩ·cm	17.6	[4]
Coefficient of linear thermal expansion	$4.8 \cdot 10^{-6}$	[3]
Refractive index (λ = 1.064 μm)	n ≈ 2.3, k ≈ 4.4	[5]
Reflectivity (λ = 1.064 μm)	R ≈ 0.84	[5]

At the same time, molybdenum as a material for metallic laser mirrors is significantly inferior to well-known metals used for this purpose, such as gold, silver, aluminum, and copper, in one key parameter – the reflectivity coefficient. Molybdenum has a relatively low reflectivity of about 60...65% in the wavelength range of 500...1050 nm [2]. As the wavelength increases, its reflectivity rises rapidly, and for wavelengths above 3 μm, the reflectivity of a molybdenum mirror exceeds 96%, reaching values of 98...98.9% (depending on polarization and incidence angle) for the 10.6 μm radiation of a CO_2 laser [6]. It is worth noting that molybdenum is also used as a material for multilayer mirrors for soft X-ray and vacuum ultraviolet radiation, applicable in astrophysics and VUV photolithography [7]. Therefore, it can be considered a promising material with a unique combination of properties for laser mirrors in the near-infrared range – particularly for laser beam control under aggressive conditions – as well as for multilayer mirrors in the spectral regions of vacuum ultraviolet and soft X-rays.

The optical quality of molybdenum mirrors depends on the method and precision of surface finishing, the purity and structure of the metal, and the melting technology. The influence of the sample's microstructure and metallurgical processing on the optical properties of the polished surface has been analyzed in several works [8, 9]. These studies confirmed that the melting technology, processing conditions, and alloy composition significantly affect the structure of the cast sample, which ultimately determines the polishing quality and the residual surface features. The correlation observed between surface roughness and microstructure indicates that smoother polished surfaces can be obtained from fine-grained samples with a high degree of texture – or, importantly, with a desired crystallographic orientation, such as in a single crystal. It should also be noted that achieving the highest optical performance requires structural homogeneity of molybdenum single crystals.

For long-distance laser beam focusing, the mirror diameter can reach a meter or more, which requires large ingots suitable for wide-format rolling. Producing large, high-quality molybdenum blanks is a challenging technological task, as traditional methods (pressing, sintering, or electron-beam melting) usually result in polycrystalline structures with anisotropic optical properties and defects that reduce reflectivity (Fernandez-Zelaia et al., 2021 [10]). Large single crystals may contain a substructure represented by sub-blocks with small crystallographic misorientations (a few degrees), which can affect laser radiation scattering and reflection efficiency.

One of the promising approaches is the formation of single-crystal blanks with controlled orientation, which minimizes scattering and interference distortions of laser radiation. Molybdenum crystallizes in a body-centered cubic (BCC) lattice, and its optical properties depend on crystallographic orientation. Reflectivity varies within 5...8% for the {100}, {110}, and {111} planes, with the maximum values observed for the {110} plane due to its higher atomic density (A. Litnovsky et al., 2023 [11]). Surface roughness, which differs among crystallographic planes even under identical processing conditions, also affects reflectivity.

Traditional crystal growth methods (zone melting, Czochralski method) do not allow for the production of large or complex-shaped crystals. In this context, the additive plasma-induction technology is of particular interest, as it combines the principles of directional crystallization with the ability to grow bulk objects layer-by-layer with a controlled geometry [12]. The application of this technology enables the production of large molybdenum single crystals in the form of plates suitable for wide-format single-crystal rolling and the fabrication of large-scale mirrors. Although this technology is actively developing for tungsten and molybdenum, its influence on the optical properties of the resulting single crystals has hardly been studied (Howard et al., 2024 [13]).

Considering the influence of crystallographic orientation on surface finish quality and the known applications of single-crystal silicon mirrors, particularly in environments with intense charged-particle fluxes in the ITER program [14], it is important to study in greater detail the effect of molybdenum crystallographic orientation on the quality of the machined surface – especially through optical radiation scattering. It is well known that the characteristics of scattered radiation, particularly laser light—such as reflectivity, the angular dependence of scattered intensity, beam divergence, polarization, and energy dependence of reflected and scattered light—are determined by the material properties and surface roughness parameters [15, 16]. Alongside electron and atomic-force microscopy, optical light scattering techniques are widely used for characterizing mirror-like metallic surfaces.

In view of the above, the aim of this work is to study the optical properties of large profiled molybdenum single crystals obtained by additive plasma-induction technology, taking into account their crystallographic orientation.

MATERIALS, EQUIPMENT, AND RESEARCH METHODS

The properties of molybdenum are provided in the Introduction. In plasma-induction growth of molybdenum single crystals, the feedstock material consists of polycrystalline rods. Their chemical composition depends on the manufacturing method and supplier. In the present experiments, technical-grade molybdenum was used, with a total impurity content not exceeding 0.05% (Table 2).

The following analytical instruments were employed to determine the chemical composition:

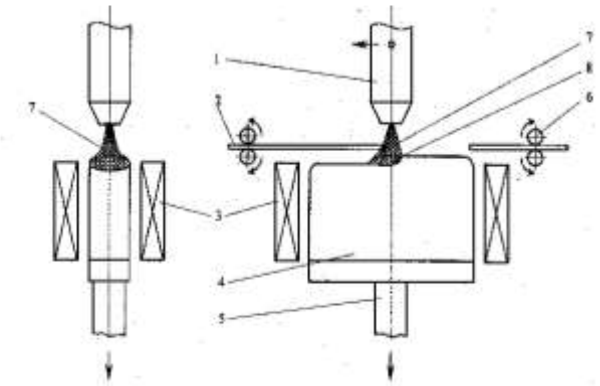
– X-ray spectrometer Philips Analytical: X'Unique II – Rh 80 kV LiF220 Ge111 TLAR;
– Carbon analyzer HORIBA EMIA-Pro;
– Oxygen and nitrogen analyzer LECO TC 436.

Table 2

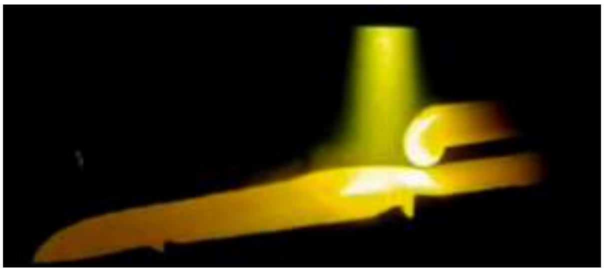
Chemical composition of molybdenum, ppm

Fe	Al	Ni	Si	C	Mg	O	N	H
20	20	30	30	30	30	15	20	5

For high-power optical components, not only the materials engineering but also the geometry and crystallographic orientation of the crystals are critical. In the case of large-diameter mirrors, the use of sizable monocrystalline plates is particularly advantageous, as it reduces labor intensity and optimizes the rolling-related technological processes. This approach also minimizes the number of processing steps and, consequently, decreases the occurrence of defects in the crystal structure. A schematic of the process and a photograph of monocrystals grown in plate form are presented in Fig. 1.



a



b

Fig. 1. Schematic and photograph of the process: a – Schematic diagram of the plasma-induction technology setup (1 – plasma torch; 2 – molybdenum feed rod; 3 – inductor; 4 – single crystal; 5 – substrate; 6 – feed rod mechanism; 7 – plasma arc; 8 – local melt pool); b – Process of single crystal growth

At the research stage, single crystals in the form of plates measuring 20×170×160 mm were used without rolling. From a crystal with, for example, a [110] orientation of the side face, samples were cut with surface orientations of [110], [100], [111], and [112]. Additionally, samples were cut from a polycrystalline billet for comparison of reflectivity.

The crystallographic orientation of the sample plates was verified using an X-ray diffractometer. An example of a diffraction pattern for one of the planes is shown in Fig. 2.

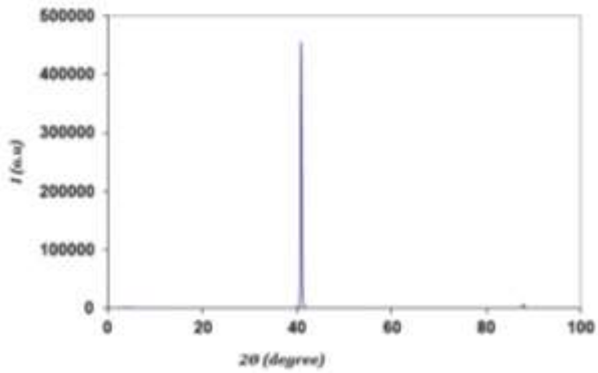


Fig. 2. Diffraction pattern of the [110] plane

The sample surfaces subjected to investigation underwent mechanical processing.

Rough grinding.

This stage was performed using abrasive wheels (corundum, silicon carbide, or diamond) with grit sizes of 80...200. The purpose was to remove excess material and form the general geometry of the samples.

Fine (precision) grinding.

This was carried out using diamond pastes with grit sizes ranging from 800 to 2000. It improved surface flatness, reduced microcracks, and prepared the surface for polishing. Oil-based cooling was applied to prevent overheating.

Polishing.

For molybdenum single crystals, special diamond suspensions or pastes (0.5...3 μm) were used on soft disks (felt or polyurethane). The goal was to achieve minimal surface roughness (Ra < 5 nm) without residual deformation. The use of colloidal silicon dioxide (SiO_2) enhanced the uniformity of material removal.

At all stages of mechanical processing, the contact pressure of the tool on the sample and the temperature were carefully controlled to prevent cracking.

DISCUSSION OF RESULTS

To investigate the optical properties of molybdenum single crystals, plates with different crystallographic orientations were grown using the plasma-induction method (Fig. 3).

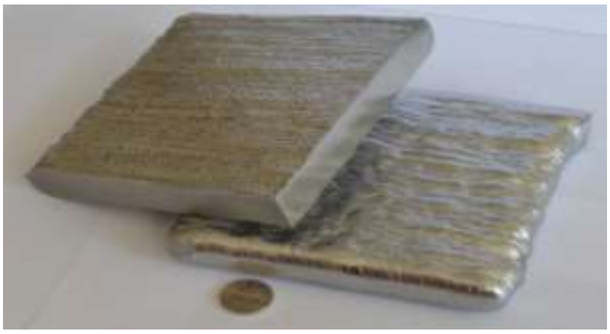


Fig. 3. Fragment of a single crystal measuring 20×150×150 mm and a molybdenum single crystal measuring 20×140×160 mm

Samples measuring 4×20×20×4 mm were cut from the single crystals. The surfaces of the single-crystal samples were processed using the unified procedure described above. The image size is 9×9 μm (Fig. 4).

To study the mirror surface of molybdenum using laser light scattering methods, a multimode helium–neon laser with a wavelength of 633 nm and a beam diameter of 4 mm was used. The laser beam was directed normally onto the mirror surface of the molybdenum single crystal, reflected from it, and projected onto a screen, where the pattern of scattered radiation was photographed. The obtained image was processed using the FIJI ImageJ software to determine the angular distribution of the scattered laser radiation.

The light scattering patterns of molybdenum single-crystal samples with orientations (110), (111), (112), and (100) (a), as well as the results of their processing in FIJI ImageJ (b), are shown in Fig. 5.

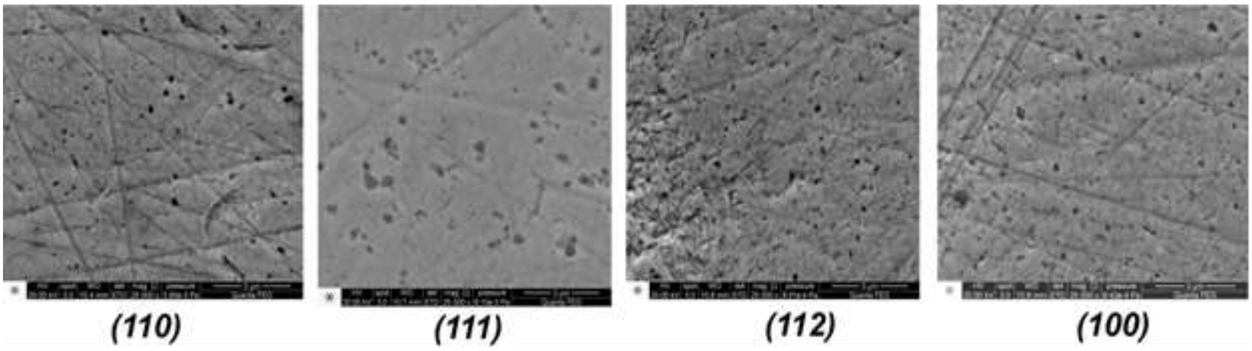


Fig. 4. Surface images of molybdenum samples (scanning electron microscope Quanta FEG, magnification ×25,000)

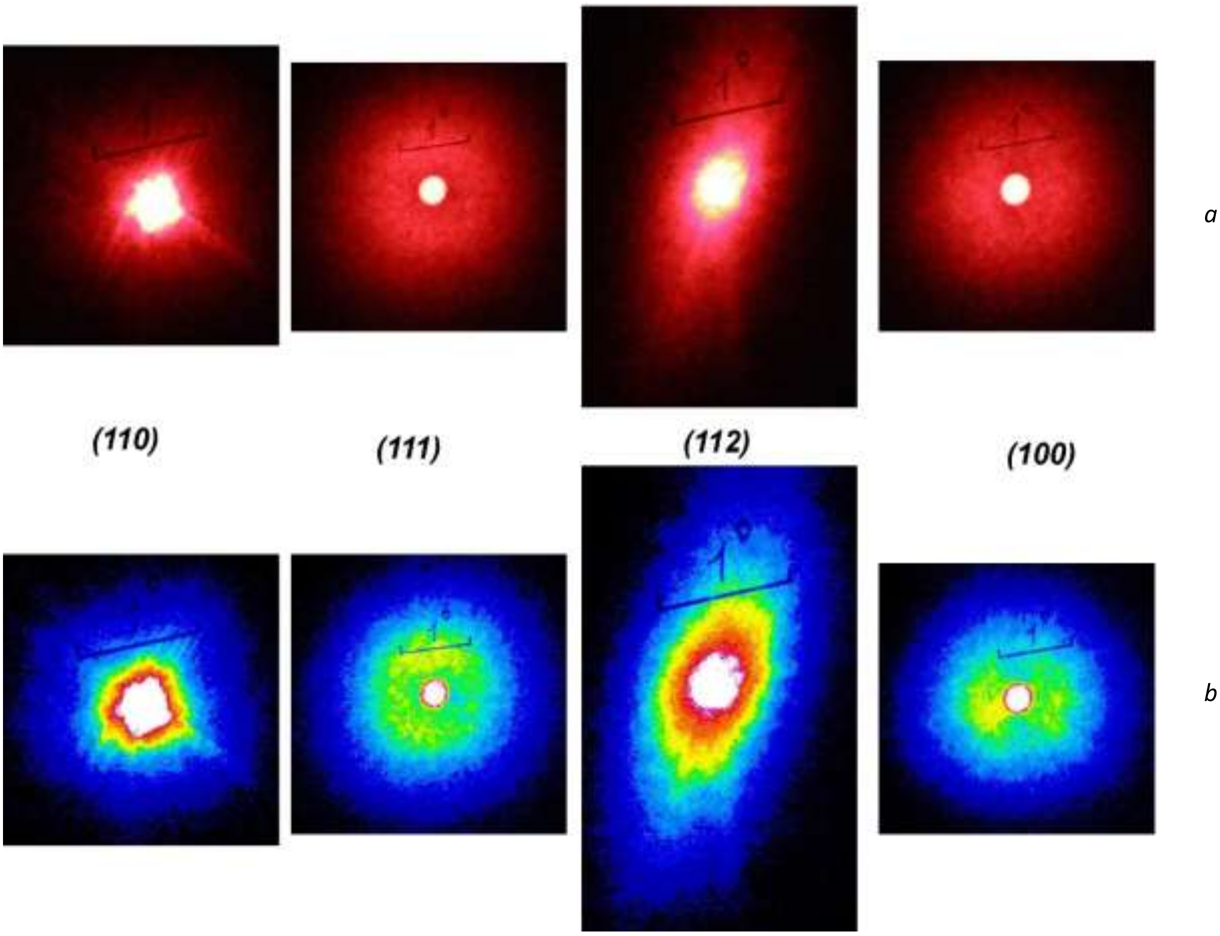


Fig. 5. Light scattering patterns from the mirror surface of molybdenum single crystals with (110), (111), (112), and (100) orientations

Diffuse scattering of laser light with a wavelength of 633 nm from different crystal planes allows for comparison of the mirror surface quality achieved by processing facets with various orientations. It is evident that the optical quality of the surface strongly depends on the crystallographic orientation. The smallest diffuse scattering angle, $2\Theta < 10°$, was observed for the (110) plane, although distortion of mirror reflection occurs at small scattering angles. For the (111) and (100) planes, the scattering behavior is similar: a clear mirror reflection pattern with a symmetric distribution relative to weak diffuse scattering at $2\Theta = 2.5°$. The (112) surface exhibits an asymmetric scattering diagram, indicating the presence of surface structural anisotropy. Clearly, this crystallographic orientation promotes the formation of a pronounced directional pattern of machining marks during polishing. The alignment of these marks along a preferred processing direction produces an asymmetric scattering pattern due to light diffraction on the surface features.

It is well known that the characteristics of scattered radiation, including laser light – such as reflectance, angular dependence of scattered intensity, beamwidth, polarization, and energy distributions of reflected and scattered light – are determined by the material properties and surface roughness parameters [15, 16]. Models linking surface topography parameters with scattered radiation characteristics are based on the effect of the surface on the phase of reflected and scattered light, as well as associated diffraction and reflection phenomena. Typically, Rayleigh-Rice and Beckmann-Kirchhoff scattering theories [17, 18], as well as the Harvey-Shack model, which characterizes scattering behavior via a surface transfer function [16], are used to predict surface scattering effects. Rayleigh-Rice vector perturbation theory shows good agreement with experimental measurements of wide-angle scattering from "smooth" surfaces for arbitrary incidence and scattering angles. Beckmann–Kirchhoff theory is valid for rougher surfaces but includes a paraxial (small-angle) assumption that limits its accuracy in handling wide-angle scattering and large incidence angles.

Analysis of the scattering diagrams allows a preliminary conclusion that the (110) plane is the most suitable for producing mirror surfaces of high-power laser optics components from large molybdenum singlecrystals grown by plasma-induction technology, despite the presence of subblock structure. This is consistent with literature data on small molybdenum singlecrystals grown by electron-beam zone melting, where the (110) plane was also identified as optimal. At the same time, the (111) and (100) planes, which demonstrate symmetric scattering patterns, could also be considered candidates for mirror surface formation when advanced surface processing techniques are applied.

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ВЕЛИКІ ПРОФІЛЬОВАНІ МОНОКРИСТАЛИ МОЛІБДЕНУ, ОТРИМАНІ ЗА АДИТИВНОЮ ПЛАЗМОВО-ІНДУКЦІЙНОЮ ТЕХНОЛОГІЄЮ, ЯК ПЕРСПЕКТИВНИЙ МАТЕРІАЛ ДЛЯ ЕЛЕМЕНТІВ СИЛОВОЇ ЛАЗЕРНОЇ ОПТИКИ

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Представлено результати дослідження великих профільованих монокристалів молібдену, виготовлених за допомогою адитивної плазмово-індукційної технології, з метою їх застосування як матеріалів для компонентів лазерної оптики високої потужності. Вибір молібдену як перспективного матеріалу для дзеркал і оптичних елементів, що піддаються термічному навантаженню, обґрунтований його високою температурою плавлення, низьким коефіцієнтом лінійного розширення, високою теплопровідністю та сильною відбивною здатністю в інфрачервоному діапазоні. Показано, що традиційні методи плавлення та формування призводять до утворення полікристалічних структур, що обмежують якість поверхні дзеркала. Запропонована адитивна плазмово-індукційна технологія, яка дозволяє вирощувати великорозмірні монокристали молібдену з контрольованою кристалографічною орієнтацією та геометрією, що придатні для виготовлення дзеркал великої площі. Було проведено порівняльне дослідження оптичних властивостей зразків із площинами (110), (111), (100) та (112) шляхом аналізу діаграм дифузного розсіювання лазера (λ = 633 нм). Встановлено, що якість поверхні дзеркала сильно залежить від кристалографічної орієнтації: найменший кут дифузного розсіювання ($2\Theta < 10°$) спостерігається для площини (110), яка є найбільш придатною для формування дзеркальних елементів високопотужної лазерної оптики. Отримані результати узгоджуються з літературними даними для невеликих монокристалів і підтверджують доцільність використання великих монокристалів молібдену, отриманих за допомогою плазмово-індукційної технології, у високопотужних лазерних системах.