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Direct laser writing of microrelief structures on chalcogenide glass by laser beam recorder of master discs

Methods to create microrelief phase elements for diffractive optics and ophthalmology were analyzed. Direct laser writing, by a laser beam recorder used for master discs is an effective method for the formation of flat microrelief phase elements on thin films of chalcogenide semiconductors, where photo-structural transformations occur under the action of laser radiation.

Key words: *direct laser writing, micro-optical elements, chalcogenide semiconductors, photo-structural transformations, laser beam recorder.*

Introduction

The application of direct laser writing (DLW) extends further than recording data on optical discs. DLW in photoresists enables the fabrication of a wide range of continuous microrelief phase elements. To improve the characteristics of optical surveillance systems, high-resolution aerial photographs require flat light-focusing elements. The elimination of aberrations to obtain clear images with a traditional lens from video surveillance systems, with high resolution, is usually achieved through the application of a large number of optical elements. This approach leads to a significant increase in the weight of the surveillance system. It is possible to solve this problem by creating small and lightweight imaging systems using diffractive optical elements [1]. Flat diffractive optical elements not only improve the performance of standard mirror-lens systems, but can also be used to create unique devices for ophthalmology. It is possible to create an artificial eye based on flat superlenses, with adaptive electrical controls for the three main parameters that determine the resulting image quality: focal length, astigmatism, and image centering. For the creation of artificial eyes, it is necessary to solve a number of complex problems, including the production of flexible flat superlenses [2].

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The technology for the DLW of various micro-optical elements requires the creation of accurate systems for rotating the substrate with a photosensitive layer, positioning the recording head according to a given law, and setting and controlling the power of the recording laser radiation. The most efficient systems can be created using circular rotation of the substrate; it allows one to create flat micro-optical elements for various purposes [3–5]. Previous studies have shown that DLW in positive photoresists can be used for the rapid fabrication of arrays of micro-optical structures (up to three times faster than a comparable electron beam lithography system) with high precision manufacturing of nanostructure elements [6]. The method of DLW of diffractive optical elements has significant advantages over other technologies for the production of phase optical elements, accurate control of process parameters, flexibility in fabricating continuous-relief micro-optical elements via a single exposure scan and development, and the ability to fabricate diffractive optical elements with arbitrary surface-relief profiles [5, 6]. The accuracy requirements for manufacturing microrelief elements and diffractive optics have increased considerably in recent years. They usually apply to the transition to a submicron and nano size range in the dimensions of the optical structure elements. The use of inorganic photoresists can alleviate some of the challenges of transitioning to the nanoscale range. Inorganic photoresists, based on chalcogenide semiconductors, are widely used in the DLW method [7–10]. Microrelief images on chalcogenide semiconductors can be obtained by various methods of DLW: local evaporation of the absorbing material, a change in the volume of the material in the radiation zone, and photo-structural transformations with subsequent selective chemical etching. Microrelief images on chalcogenide semiconductor films can be formed under the action of laser pulses of various durations. Photo-structural transformations in chalcogenide semiconductors allow one to create multi-level microrelief images. Industrial laser beam recorders were developed for the DLW of information on master discs, with a layer of positive photoresist. Those are characterized by the rotation of the substrate at a predetermined, constant linear rate and accurate radial movement of the recording head. The possibilities of using a standard laser beam recorder of master discs for the DLW of diffractive optical elements over a large area, and micro-optical elements, were analyzed.

Direct laser writing of phase optical elements

The technology to create flat micro-optical elements of variable resolution was developed more than twenty years ago. There were two main approaches for their development: a method to create blazed binary gratings and the «echelette» method for blazed gratings. The disadvantage of the latter approach is the limitation owing to the shadowing effect. In both cases, the height of the patterned structures is typically equal to the wavelength of the incoming light, which makes the manufacture of micro-optical elements for large surfaces technologically complex [11]. The technology to form flat microlenses is based on photolithography methods, namely, the application of photoresist layers, laser exposure, selective etching of the photoresist layers, and the substrate application [12]. Therefore, together with the methods to laser record microelements, the technology to diamond-cut microlenses was developed. The use of mechanical material removal to create micro-optical elements is difficult because it requires the use of precise machine tools and expensive diamond cutting tools [13, 14]. An alternative

method to develop flat micro-optical elements is by replacing standard, bulky, and expensive lenses with a high numerical aperture, consisting of a large number of lenses. Excessive weight and size limit the use of a multi-lens in compact and cost-effective optical systems. However, flat lenses with a high numerical aperture can be widely used in video surveillance systems, microscopy, and small-sized spectrometers. Achieving a high numerical aperture in flat lenses, using the diffractive optical elements of the components, is a difficult task. This is due to phase distortions because their constituent structures are of a wavelength scale [15]. The problem of eliminating aberrations and phase distortions, when creating high-aperture optics based on plane diffractive optical elements, can be solved by using many level elements, the number of which can be several hundred [1, 16]. The development of diffractive optical elements with 256 levels was reported [1]. The spatial resolution of the process of creating micro-optical elements using DLW is primarily determined by the spot size of the focused laser beam. This subsequently depends on the wavelength and characteristics of the recording materials, and on the accuracy of the two-coordinate positioning systems used in the recording process [7].

The formation of nanosized relief on chalcogenide semiconductors by a laser beam recorder of master discs

Unlike conventional high refractive index glasses, chalcogenide glasses can undergo distinct radiation-induced structural modifications, at comparatively low power densities and exposure doses [7]. Chalcogenide glasses have gained extensive interest, not only because of their high refractive index, wide transparent window, and high optical nonlinearity, but also for their rich physicochemical properties, such as phase changing and photopolymerization, upon light exposure [17, 18]. These unique properties have made chalcogenide glasses an excellent candidate for diverse applications, including micro-optical elements and metamaterials [17, 19]. The processes that are based on photo-structural transformations in chalcogenide glassy semiconductors satisfy the most requirements for the technology required to form flat optical elements and microrelief phase elements. The microrelief images on chalcogenide semiconductor films can be formed under the action of laser pulses of different durations. Photo-structural transformations in chalcogenide semiconductors facilitate the creation of multi-level microrelief images that allow the acquisition of flat, high-performance, focused microelements. One of the substantial advantages of using inorganic photoresists, based on chalcogenide semiconductors, is the possibility to use laser recording for bumps (on a negative resist) or pits (on a positive resist) of various height (depth). Inorganic resists are characterized by a wide dynamic range [8, 20]. Typical exposure characteristics of an inorganic resist are shown in Fig. 1 [20].

Experimental studies on the process of recording data on a positive inorganic resist, using the laser beam recorder of master discs, showed that it is possible to form pits of specific sizes. The depth of the pits was determined by the thickness of the resist and length/width of the pits, by the recording laser power and the chemical etching mode [8]. Nickel copies were made of the original discs, which were used to replicate the CDs. Recording data on a layer of negative inorganic resist, deposited on a nickel substrate, made it possible to obtain relief structures after selective chemical etching, by which relief copies on polycarbonate were obtained by thermal pressing. Relief struc-

tures on the inorganic resist layer were shown to have a sufficiently high mechanical strength, which allows the formation of diffractive micro-optical elements directly in the resist layer, without additional processing of the substrate material [21]. The resolution of inorganic photoresists, based on chalcogenide semiconductors, allows recording tracks with a width of 0,3–0,5 microns. The minimum track width recorded by the laser beam on the films of the inorganic resist is primarily determined by the resolution of the optical system, which focusses the laser recording radiation [22]. The images of the tracks recorded on the positive and negative inorganic resists are shown in Fig. 2 and Fig. 3, respectively.

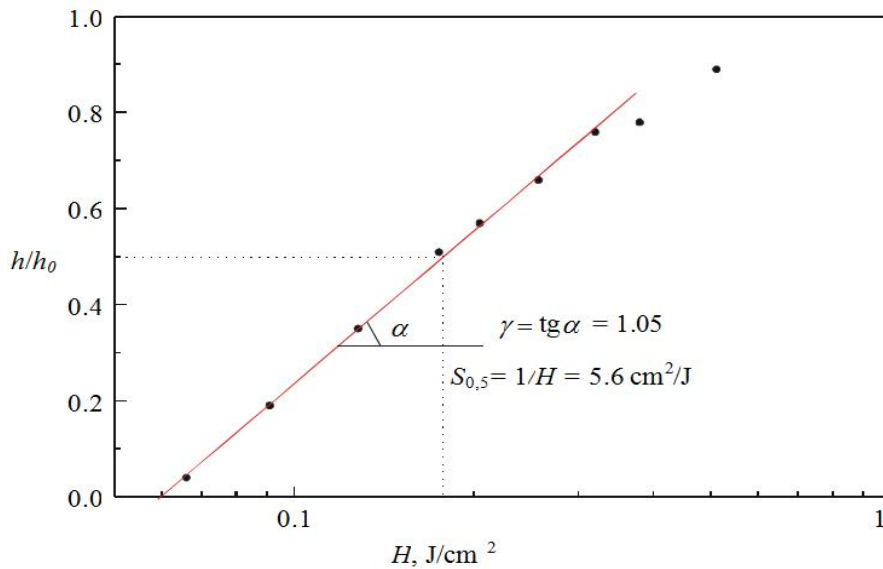


Fig. 1. Characteristic curve of the inorganic negative resist As_2-S_3 [20]

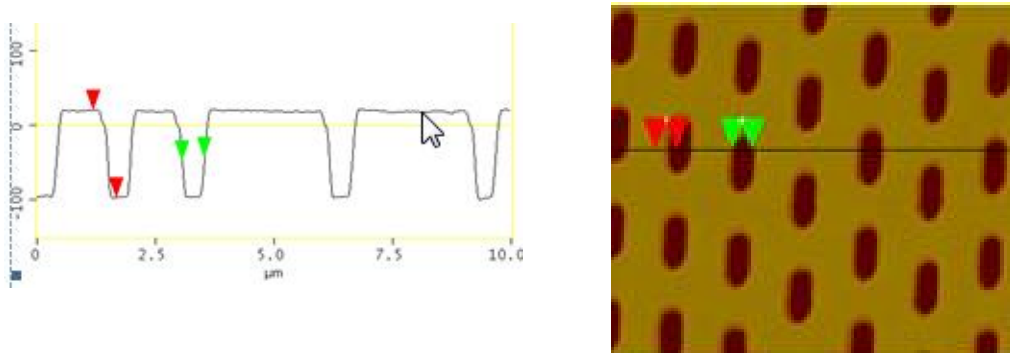


Fig. 2. The relief microstructure on the positive inorganic photoresist $GeSe_2$. [23]

On inorganic resists, it is possible to obtain both negative images (on As_2S_3 resists) and positive images (on $GeSe_2$ resists). The type of resist depends on both the composition and the selective etching used [8]. To obtain micro-optical elements by direct laser recording, chalcogenide semiconductors of various compositions can be used: As-based glasses ($As_{35}S_{65}$) and Ge-based glasses for the formation of images in the UV-range (244 nm) [7]; 3D micro-optical elements formed by the photo-structural

transformations of germanium silicate by DLW [25]; telluride (PbTe) chalcogenide for the formation of micro-optical elements for the near IR-range [26]. Using DLW with femtosecond pulses of UV-radiation, diffraction gratings were recorded on thin films of As_2S_3 [17]. Nanostructured arrays were photo-patterned on As_2S_3 films using multiphoton DLW [27]. Combined elements of various heights can be performed by exposing the negative resist to laser radiation of various power [24].

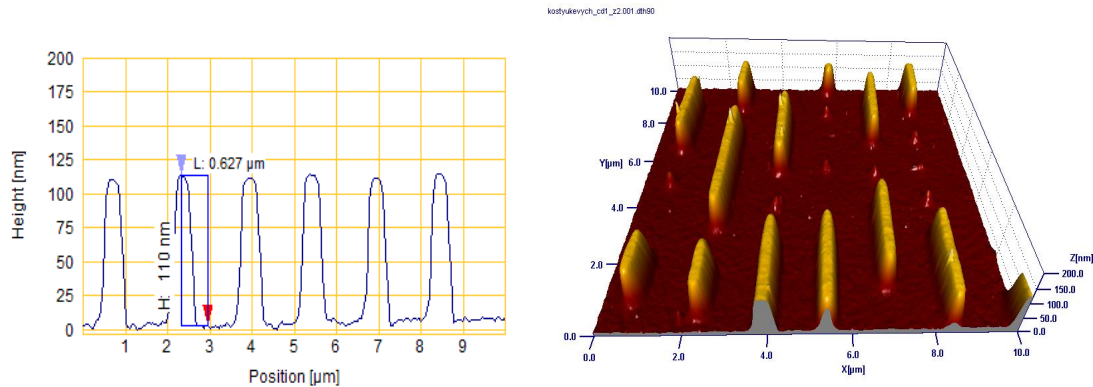


Fig. 3. The relief microstructure on the negative inorganic resist As_2S_3 [24]

Using the laser beam recorder of master discs to form micro-optical elements

For the manufacture of microrelief structures and diffractive optical elements, there are many non-standard technologies that use sophisticated modern equipment [28–30]. Research on the development of circular laser recording systems for the production of micro-optical elements began more than 20 years ago. It was previously shown that a micro-image can be formed by means of a continuous rotation of the substrate, while the focused light beam is simultaneously moved along the radius of the substrate. Such systems provide a recording of microrelief images at high speed. It is possible to produce a mask of specified configuration, using a computer-controlled amplitude modulation of the laser beam [28].

In recent years, a new generation of laser recording devices was developed, which can be used for the manufacture of micro-optical elements. A standard laser beam recorder of master discs, with a modified laser beam control system, can be used to create Fresnel lenses, diffraction optical elements and micro-optical elements, using the DLW method. Laser beam recorders of master discs facilitate the recording of tracks with a given constant pitch, with a minimum radial runout of focused tracks on a photoresist. Micro-optical elements should be written by a laser beam recorder of master discs during continuous spiral scanning and circular raster scanning processes. In this case, the scanning of a material surface is fulfilled by two types of functional element movements: substrate rotation and radial displacement of the recording beam. This method provides the maximum writing speed for micro-optical elements [28]. A standard laser beam recorder of master discs has an autofocus system, with an electrodynamic or piezoelectric actuator that supports the focusing plane and the substrate during the writing process, with an accuracy of $0,05 \mu\text{m}$. To ensure minimal radial beats, a laser recording station uses an aerostatic spindle with a beat radius of no more than $0,1 \mu\text{m}$. DLW by

laser beam recorder allowed the fabrication of multiphase levels in one photolithography process step [31].

It is of considerable interest to use such high-precision equipment for the manufacture of micro-optical elements. Circular scanning laser recording systems which include the laser beam recorder of master discs allow making the formation of diffraction optical elements for large areas. The radial beat of the tracks recorded on the laser recording station does not exceed 50 nm. The modification of the control system of the laser beam recorder of master discs allowed the recording of tracks of different depths (including different depths of the elements of one track) by controlling the recording power. To obtain tracks of different widths, recording should be performed by the sequential exposure of tracks, with a recording head displacement of the width of the track. The recording of individual tracks can be emitted with radiation of varied power, which allows the formation of images with a given profile. Micro-optical elements were recorded at a linear velocity of 2,4 m/s with a 405 nm laser beam, which was focused into a spot with a diameter of 0,5 μm . The minimum width of the recorded lines was 0,5 μm . Binary optical elements can be recorded with a laser beam recorder. A diagram of the process of laser recording binary optical elements with inorganic resists is shown in Fig. 4. They are created in a photoresist layer by sequentially exposing annular zones of various widths. The microprocessor control system of the station should be able to record zones of a given width by exposing a different number of contiguous tracks. After selective etching of the photoresist, it can etch the substrate material to a predetermined depth.

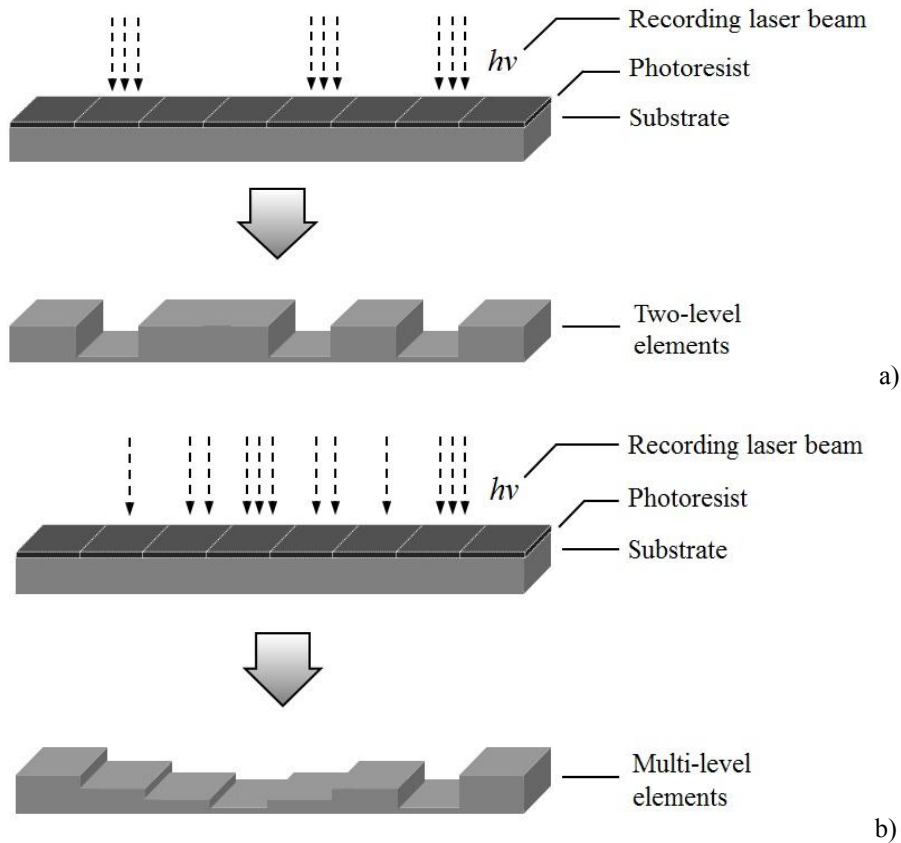


Fig. 4. Creation of diffraction optical elements on an inorganic resist:
a) two-level elements; b) multilevel elements

Our experimental studies showed that laser recording of diffraction elements with a high linear velocity (2,4 m/s) without local thermal destruction is possible on chalcogenide glasses of the AS-S-Se system. As shown during the recording of two-level optical elements on an inorganic resist by the laser beam, recorder zones of different widths were formed by radiation, with a laser beam of constant power at different numbers of passes. However, the process of manufacturing multilevel elements is more complicated. During multilevel optical elements laser beam recording on inorganic resists, zones of various widths and heights have to be written with different laser recorder radiation powers.

Conclusion

1. DLW facilitates the manufacturing of multi-level diffraction optical elements with an arbitrary relief profile.

2. Standard laser beam recorders of master discs can be used as circular scanning laser systems for the direct laser recording of diffractive optical elements and micro-optical elements on photoresist layers. The laser beam recorder of master discs with circular scanning is a universal tool for the fabrication of diffractive optical elements. For laser recording of diffraction and micro-optical elements, a monitoring system for the positioning of the recording head is necessary. Future development should allow for the recording of tracks of various widths, with a given variable pitch, and recording with different exposure levels in each zone.

3. In direct laser recording for the formation of diffractive optical elements, inorganic photoresists based on chalcogenide semiconductors can effectively be used. These semiconductors have a high refractive index, a wide spectral range of transparency, and allow the formation of images on which various photo-induced processes are used. Laser recording of diffraction elements with a high linear velocity (2,4 m/s) without local thermal destruction is possible on chalcogenide glasses of the As-S-Se system.

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1. Nikonorov A., Petrov M., Bibikov S., Yakimov P., Kutikova V., Yuzifovich Y., Morozov A, Skidanov R., Kazanskiy N. Toward Ultralightweight Remote Sensing With Harmonic Lenses and Convolutional Neural Networks. *IEEE Journal of selected topics in applied earth observations and remote sensing*. 2018. P. (99):11.

2. She A., Zhang S., Shian S., Clarke D., Capasso F. Adaptive metalenses with simultaneous electrical control of focal length, astigmatism, and shift. *Science advances*. 2018. 4. No. 2. Article number Eaap 9957.

3. Gale M. Fabrication of continuous-relief micro-optical elements by direct laser writing in photoresists. *Optical Engineering*. 1994. 33. P. 3556–3564.

4. Hafner M., Pruss C., Osten W. Laser direct writing. Recent developments for the making of diffractive optics. *Optik & Photonik*. December 2011. No. 4. P. 40–44.
5. Mohammad N., Meem M., Shen B., Wang P., Menon R. Broadband imaging with one planar diffractive lens. *Scientific Reports*. 2018. **8**. Article number 2799.
6. Braun A., Maier S. Versatile Direct Laser Writing Lithography Technique for Surface Enhanced Infrared Spectroscopy Sensors. *ACS Sensors*. 2016. **1**. P. 1155–1162.
7. Schroeter S., Vlček M., Poehlmann R., Fišerová A. Efficient diffractive optical elements in chalcogenide glass layers fabricated by direct DUV laser writing. *Journal of Physics and Chemistry of Solids*. 2007. **68**. P. 916–919.
8. Petrov V., Kryuchin A., Kostyukevich S., Rubish V. Inorganic Photolithography. Kyiv: Institute for Physics of Metals, NAS of Ukraine, 2007. 196 c.
9. Almeida J., Paula K., Arnold C., Mendonça C. Sub-wavelength self-organization of chalcogenide glass by direct laser writing. *Optical Materials*. 2018. **84**. P. 259–262.
10. Stronski A.V., Schepeljavi P.E., Kostyukevych S.A., Romanenko P.F., Robur I.I., Indutnyi I.Z. Fabrication of Fresnel lenses and other optical elements with the help of inorganic resists. *Proc. SPIE*. 2213. Nanofabrication Technologies and Device Integration (28 July 1994); <https://doi.org/10.1117/12.180953>
11. Paniagua-Dominguez R., Yu Y.F., Khaidarov E., Choi S., Leong V., Bakker R., Liang X., Fu Y., Valuckas V., Krivitsky L., Kuznetsov A. A Metalens with a Near-Unity Numerical Aperture. *Nano Letters*. 2018. **18**. P. 2124–2132.
12. Swanson G. Binary Optics Technology: The Theory and Design of Multi-level Diffractive Optical Elements. URL: <https://apps.dtic.mil/dtic/tr/fulltext/u2/a213404.pdf>
13. Jasinevicius R., Pizani P., Cirino G.A. Ultraprecision machining of diffraction optical elements on soft semiconductor crystal. *The International Journal of Advanced Manufacturing Technology*. 2015. **77**. 1154–1163.
14. Kryuchyn A. High-precision micro-prism structures and prospects for their large-scale application. *Bulletin of the National Academy of Sciences of Ukraine*. 2018. **4**. P. 45–53.
15. Khorasaninejad M., Chen W., Devlin R., Oh J., Zhu A., Capasso F. Metalenses at visible wavelengths: Diffraction-limited focusing and subwavelength resolution imaging. *Science*. 2016. **352**. P. 1192–1194.
16. Nikonorov A., Bibikov S., Myasnikov V., Yuzifovich Y., Fursov V. Correcting color and hyperspectral images with identification of distortion model. *Pattern Recognition Letters*. 2016. **83**. Part 2. P. 178–187.
17. Zhang Q., Lin H., Jia B., L. Xu, M. Gu. Nan gratings and nanoholes fabricated by direct femtosecond laser writing in chalcogenide glasses. *Optics express*. 2010. **18**. 6885.
18. Zakery A., Elliott S.R. Optical properties and applications of chalcogenide glasses: a review. *Journal of Non-Crystalline Solids*. 2003. **330**. P. 1–12
19. Kostyukevitch S., Shepeljavi P., Indutnyi I., Stronski A., Stetsun A. Formation of optical disk direction paths and optical master disks with the help of inorganic resists. *Proc. SPIE 2026, Photonics for Processors, Neural Networks, and Memories* (9 November 1993); doi: 10.1117/12.163608.
20. Kostyukevych S., Shepeliavij P., Svechnikov S., Moskalenko N., Tomchuk V., Koptyuh A., Volkov A., Kazansky N., Kostyuk G. Formation of diffractive optical elements using inorganic laser lithography. *Data Recording, Storage and Processing*. 2002. Vol. 4. No. 3. P. 3–12.
21. Kostyukevych S., Shepeliavij P., Moskalenko R., Wenger V., Petrov V., Kryuchyn A., Shanoilo S. The study of the mastering of compact discs on inorganic photoresists. *Data Recording, Storage and Processing*. 2001. Vol. 3. No. 4. P. 5–14.
22. Kryuchyn A., Petrov V., Rubish V., Trunov M., Lytvyn P., Kostyukevich S. Formation of anoscale Structures on Chalcogenide Films. *Physica status solidi (b)*. 2017. **255**. Article number 201700405. DOI: 10.1002/pssb.201700405.
23. Indutny I., Kryuchin A., Borodin Yu., Danko V.A., Lukaniuk M.V., Minko V.I., Shepeliavij P.E., Gera E.V., Rubish V.M. Optical Recording of Micro- and Nano- Relief Structures on Inorganic Resists Ge-Se. *Data Recording, Storage and Processing*. 2013. Vol. 15. No. 4. P. 3–14.
24. Kryuchyn A., Petrov V., Rubish V., Lapchuk A., Kostyukevych S., Shepeliavij P., Kostyukevych K. High-speed optical recording in vitreous chalcogenide thin films. *Semiconductor Physics, Quantum Electronics & Optoelectronics*. 2014. **17**(4). P. 389–393.

25. Malinauskas M., Zukauskas A., Purlys V., Gaidukeviciute A., Balevicius Z., Piskarskas A., Fotaki C., Pissadakis S., Gray D., Gadonas R., Vamvakaki M., Farsari M. 3D microoptical elements formed in a photostructurable germanium silicate by direct laser writing. *Optics and Lasers in Engineering*. 2012. **50**. P. 1785–1797.
26. John-Wallace. Metasurface Optics: Broadband achromatic metalens focuses all polarizations. URL: <https://www.laserfocusworld.com/home/contact/16572305/>
27. Schwarz C., Williams H., Grabill C., Lewis A., Kuebler S., Gleason B., Richardson K., Pogrebnyakov A., Mayer T., Drake C., Rivero-Baleine C. Processing and fabrication of micro-structures by multiphoton lithography in germanium-doped arsenic selenide. *Proc. of SPIE*. 2014. **8974**. 89740P–1–6.
28. Koronkevich P., Kiryanov V., Korol'kov V., Poleshchuk A., Cherkashin V., Churin E., Kharissov A. Fabrication of diffractive optical elements by laser writing with circular scanning. *Proc. SPIE*. 1994. **2363**. P. 290–298.
29. Perlo P., Sinesi S., Ripetto M., Uspleniev G. The use of a circular laser recording system for the manufacture of halftone patterns of diffractive optical elements based on DLW glass plates. *Computer Optics*. 1997. **17**. P. 85–97.
30. Poleshuk A., Korolkov V., Bessmeltsev V., Nikonorov Yu., Karvan A., Verkhogliad A. Precision laser technological complex for the production of scales, grids, photo masks and synthesized holograms based on laser three-dimensional micro- and nano-processing. <http://www.holography-journal.com/wp-content/uploads/2013/03/071.pdf>. P. 66–72.
31. Wang Y., Zhang W., Yang Z., Xiong X., Xia L., Gao M., D. Zhang, Wang D., Yuan J. Fabrication of large diffractive optical elements by laser direct writing IEEE International Conference on Manipulation, Manufacturing and Measurement on the Nanoscale (3M-NANO) (18-22 July 2016, Chongqing). Chongqing, 51–54. 2016. DOI:10.1109/3M-NANO.2016.7824973.

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