ReviewPaper

Infrared chalcogenide microlenses

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The state –of-art in the filed of infrared chalcogenide microlenses is presented. The development of the infrared kinoform optical elements based on glassy chalcogenides, down to nanometric size, is discussed. The advanced methods of making lenslets and lenslet arrays are reviewed.

1. Introduction

The advances in optoelectronic devices are based on the achievements in optics, electronics and last but not least in material science. Among the most important materials used in optoelectronics are the amorphous chalcogenide materials. The chalcogenides are those materials that contain one or more of the chalcogen elements: sulphur, selenium, tellurium [1].

Amorphous and glassy chalcogenides are suitable for applications due to their special properties in relation to the light including infrared transparency and modification of the refractive index [2-4]. In the recent years the research in non-crystalline chalcogenides increased tremendeously, numerous publications being related not only to experimental [5-61] and theoretical [62-72] features of various chalcogenides, but also to applications [73-84].

Chalcogenide glasses are characterized by high values of the refractive index in the range 2.3-3.2 and, therefore, the chalcogenide optical elements can be used for coupling IR light beams into chalcogenide glass waveguides.

Micro-prism and micro-mirror arrays are important elements of modern electro-optics, used for several purposes such as optical computing, optical communications, beam coupling or beam combination, for integrated planar optical interconnections, etc.

Microlenses and microlens arrays can be found in many optoelectronic applications, such as optical communication and computing, CCD cameras, faxes, imaging systems and IR technology [85].

Micro-optical lenslets are considered in free-space optical systems providing an optical interconnection between very large scale integrated (VLSI) electronic chips [86], between modules containing chips [87], and in stacked planar optics [88].

Optical elements, the fabrication of kinoform of chalcogenide lenslets and lenslet arrays are reviewed.

2. Kinoform optical elements and chalcogenide lenslets

The kinoform is a phase-modifing object. The kinoform lens reprezents one of the best opportunities for nanometer-scale spot sizes used e.g. for hard X-rays.

The modification of the refractive index in chalcogenide films under the action of the electromagnetic radiation was used for the production of optical elements with thin film phase and elements of integrated optics, such as kinoform elements. The kinoform lenses are refractive lenses with all material causing redundant 2π phase shifts removed to reduce the absorption problems inherently limiting the resolution of the refractive lenses. By stacking kinoform lenses together, the effective numerical aperture, and thus the focusing resolution, can be increased.

Arsenic trisulfide glass has some very unique properties. The light red glass transmits from the visible out to 8 μm with no appreciable absorption. For systems operating in the near infrared or in the 3–5 μm window, the glass is extremely useful for lenses or windows. Generally, the use in the 8-12 μm range is not recommended because of intrinsic absorption. Arsenic trisulfide glasses has the lowest thermal change in refractive index of any infrared optical material in use today including germanium, zinc selenide and zinc sulphide. For this reason, lenses or windows made from As_2S_3 glass do not show optical distortion when subjected to the intense IR radiation from lasers such as YAG, Er:YAG, or CO_2.

In order to record kinoform optical elements As_2S_3 films with the thickness 6-8 μm were used. Such a thickness is optimal for a phase shift up to 2π in the irradiated regions when the recording is performed by an Argon laser and the reading by a He-Ne laser [89]. The films were prepared by electron-beam evaporation. In chalcogenides have been recorded kinoform cylindrical lenses. A complicated mask consisting of twelve different triangles was used in this purpose. An array of kinoform cylindrical lenses has been recorded. It was possible to reproduce kinoform optical elements in high amount, to make spatial phase filters, aspherical lenses and other optical elements, which are produced by mechanical treatment of glass with difficulty. It was possible to produce waveguide lenses, geodesic lenses and Lundberg lenses.

The geodesic lens is a non-planar part of the waveguide with the shape of a cupola. Lundberg lens is a three-dimensional lens where the refractive index increases smoothly from the index value of the waveguide

border to a maximum value in the center of the lens. The creation of these lenses is a difficult task. The advantage of the chalcogenide glasses is the possibility to use the laser for the local modification of the optical parameters. The Lundberg lenses were obtained by deposition through a diaphragm of variable radius. The correction of the lens needed in this case is carried out by exposing the border part of the lens to0 a laser radiation. The holography can be used in the fabrication of Fresnel lenses based on chalcogenide materials, with high transmission in infrared spectrum, low level of stray light (10^{-6}) and diffraction efficiency as high as 85 % [90].

The possibility to make microlenses was demonstrated. The chalcogenide glasses can be shaped under light illumination [91]. The fluidity of the glass is increased by illumination and, under external forces, the shape of the material can be changed.

Micron sized lenses or lenslets have been fabricated in a variety of ways. Ishihara and Taginaki [92] have shown lenslet formation by melting resins deposited on top of a CCD image sensor but precise control of the feature size was cumbersome. A variation of this technique by Popovic et al. [93] involved a multistep process consisting of metal deposition, photolithographic definition of apertures and then melting the deposited resin. Lenslets have also been demonstrated [94] in InP by first chemically etching a multilevel MESA and then using mass transport to get a smooth curvature.

Saitoh et al. [95] have shown that a photolithographic technique using chalcogenide glasses is promising for preparation of automatically positioned microlenses on fiber-end centers. The principle is demonstrated by use of a typical chalcogenide glass, As_2S_3.

Microlenses were fabricated on end surfaces of single-mode silica fibers with a core diameter of 6 μm and a cladding diameter of 125 μm as follows: First, As_2S_3 film was deposited by thermal evaporation onto the flat end surfaces of the fiber under pressure of 10^{-3} Pa, with a deposition rate of ~3 nm/s. The typical film thickness was 1 μm. The film was exposed to a He-Ne laser beam (λ = 633 nm). Finally the film was etched in NaOH several minutes. The etching of the exposed a unexposed parts of the films is different. Thus does appear a spherical convex structure, which replicates a Gaussian-like intensity distribution of exposed light over the fiber core. The focusing operation of the convex structure was clearly demonstrated by the authors. The spot diameters evaluated at the light intensity of 1/e of the central intensity are 5.9 μm for the fiber surface and 2.9 μm at the 10 μm distance.

The shape of the convex structure and the focusing characteristic correspond quantitatively. Geometric optics suggest that the focal length f of a spherical plano-convex lens is approximately

$$f \approx \frac{r^2}{2(n-1)d},$$ (1)

where 2r is the lens diameter, n is the refractive index of the lens, and d is the lens thickness. Putting 2r = 4.5 μm,

n = 2.6, and d = 80 nm, one gets f = 20 μm. This f value is longer than the value (~10 μm) that was experimentally measured, and the difference probably is due to the rough evaluation procedure, which neglected the precise lens shape, aberration, and guided-wave and diffraction effects.

In this method the lens is automatically positioned on the fiber core, and the lens diameter becomes nearly the same as the core diameter. Then, the thickness and shape of the lens are crucial and are determined by three factors: the initial film thickness, the light exposure, and the etching process.

Other chalcogenide glasses, as e.g. As_2Se_3, can be used for the same purpose: to focus the light emerging from the fibers.

Ramachandran et al. [96] have shown that it is possible to use photo-expansion in order to write optically lenslets in As_2S_3 glass. The key advantage of this technique is that it is a one-step optical process, which requires no multistep etching or fixing to get a smooth curvature. Illumination of chalcogenide glasses with above band-gap light causes photodarkening and photo-expansion. Photodarkening is a photo-induced red-shift of the optical absorption edge which is accompanied by an increase in the index of refraction in the transparent spectral range below the absorption edge. Photo-expansion is the attendant increase in the lattice volume of a photodarkened chalcogenide glass. As_2S_3 expands approximately 0.5 % due to the photostructural change. However, when the irradiated volume is constrained in an unexposed matrix, stress at the interface leads to higher volume changes. Hisakuni and Tanaka [97] obtained a 2 % giant expansion by illuminating a part of an As_2S_3 sample with 632.8 nm light from a He-Ne laser. They used this effect to fabricate lenslets and lenslet arrays with H-Ne laser.

Assuming the lenslets are spherical in profile, the focal length may be calculated by:

$$f = \frac{h^2 + \dfrac{d^2}{4}}{2h(n-1)}$$ (2)

where h is the height, d is the diameter of the lenslets, and n is the index of refraction of the glass. For 1550 nm light, n = 2.4380 for As_2S_3 glass, giving a focal length f = 770.3 μm for the lenslet exposed for 5 min. and a focal length of 369.5 μm for the lenslet exposed for 15 min. For the lenslet induced by a 5 min exposure the observed focal length is smaller than the value predicted, whereas the lenslet written for 15 min has a longer focal length than the model predicts. The authors explain the discrepancy with the experimental results by considering the mechanism of the photodarkening process, which produces an attendant refractive index change beneath the surface of the lenslets of As_2S_3. The temporal dynamics of the index change are non-linear and thus the total index change for a 15 min exposure is much larger than for a 5 min exposure. Thus, in the case of the lenslet exposed for 15 min., the focusing effect is due to a combination of the

relief structure on the surface as well as a radial gradient index distribution within the glass matrix, while the model considers only a surface modulation. On the other hand, the contribution due to the index distribution for the 5-min lenslet is negligible. The absolute departure from the predicted values for focal lengths in both cases may arise primarily from a Gaussian-shaped profile being modeled as a hemispherical lenslet. Also, the fact that lenslets are not hemispherical in profile may explain the reason for the absence of complete collimation.

Semiconductor lasers have been widely employed in many kinds of optoelectronic devices, despite the inconvenient elliptical beam cross-section of edge-emitting lasers. Efficient coupling to conventional lenses and fibers requires a circular beam cross-section, which has been obtained by use of aspherical microlenses or combination of spherical and cylindrical lenses. However, for alignment and fixing of a microlens to a laser surface, sophisticated techniques and stringent manipulations are required. In addition, it is difficult to assemble compactly the laser-lens system. A principle of self-developing microlenses for semiconductor lasers has been demonstrated by Saitoh and Tanaka [97] using the photosensitive glass As_2S_3. The glass film is placed in front of a semiconductor laser, the light from each increases the refractive index and the thickness of the film. If its position and thickness are properly selected, the illuminated film operates, as an aspherical microlens which can focus an elliptical laser beam to a circular spot. No lens alignement is needed and the laser-lens assembly can be compact.

Chalcogenide films are promising materials for producing passive elements of integrated optics. Directed light waveguides, prisms, lenses, and so on, can be recorded by a laser or electron beam on them [98].

On the other hand, chalcogenide lenses got by molding are important for infrared optical systems, such as thermal imaging and night vision systems. Currently, most chalcogenide glasses (based on selenium) are produced as small boules in sealed quartz ampoules, which help to minimize the loss of selenium which is prone to evaporate due to its relatively high vapor pressure. Once the glass is formed, any practical application requires that the glass be transformed into an optical component, such as a lens. Current production processes for such optics rely mainly on grinding or molding the chalcogenide glasses into lenses. These, however, are rather lengthy processes. After the glass is formed from its constituent elements in quarz reaction container, the glass is cast onto a plate, and annealed so as to avoid breakage. The annealed plate is then cut into blanks, which are ground to thickness, edged, and turned or ground into lenses or formed into lenses in a vacuum press. This process can take several days to complete. Furthermore, the lenses are constrained by the size of the mold ovens used to produce them. Various compositions have been used to produce chalcogenide lenses, for example those based on ternary and quaternary glasses based upon selenium (Ge-Sb-Se and/or Ge-Sn-Sb-Se) Typical members are: $Ge_{12}Sn_7Sb_{13}Se_{68}$, $Ge_{28}Sb_{12}Se_{60}$, In reacting the constituents to form a chalcogenide glass, the elemental constituents are loaded into a quartz reaction reactor. Typically, the constituents are layered in the container from top to bottom as follows: Sb, Se, Ge, Se, and Sn. Notice that the layer of germanium is disposed between layers of Se, the two layers dividing between them the total weight of selenium in the reaction melt. The glass, once formed, is cast, molded or machined into shape. Casting typically involves pouring the liquid glass into a mold, and molding typically involves placing a sold preform into the mold. Each mold half defines the shape of one face of a lens. A taper and shoulder on each mold half provide precise control of centering and perpendicularity of the two mold halves. The construction material of the mold must be carefully selected. As the chalcogenide lens and mold expand and contract as they are heated and cooled, it is important to select a mold material that has a coefficient of thermal expansion similar to that of the chalcogenide glass being formed. Otherwise, as the lens and mold expand and contract at different rate, the lens could potentially be damaged. A material with acceptable thermal expansion coefficient is hardened 420 stainless steel. Because the chalcogenide material can erode the tool steel up to a 1/8 inch deep in one casting cycle, the mold must be coated with a surface coating that protects the mold surfaces and yet not degrade the optical surfaces of the lens. An example is titanium nitride (TiN).

An optically athermalized infra-red lens having a group of three closely spaced lens elements has been produced [Michael Roberts, Patent 4,679,891, 1987, USA]. The group is of positive overall power. The lens elements are made of different infrared transmitting materials. Each has a positive thermal coefficient of refractive index. Two of the lens elements are made of materials whose refractive indices are relatively temperature insensitive, one, e.g. of zinc selenide or chalcogenide glass, being positively powered and the other, e.g. of zinc sulphide, being negatively powered to effect achromatization and correct spherical aberration. The third element is negatively powered and of a relatively temperature sensitive refractive index material, e. g. germanium, of lower dispersion and higher refraction index than the other two. This complex lens is used as objective of an afocal telescope having an eyepiece and associated scanner and detector arrangement and may effectively athermalize the whole system. Another version of the complex infrared lens consists in a positive group of four closely spaced lens elements. The front element is a negative meniscus convex to the front of zinc sulphide, the next is a positive biconvex element made of IRG 100 (Schott) chalcogenide glass, the next is a negative bi-concave element of germanium, and the back element of the group is a positive bi-convex element of IRG 100 chalcogenide glass. The objective lens has a rear component formed by three lens elements of which the front element is a positive bi-convex element of germanium, the middle element is a negative bi-concave element of BS1 chalcogenide glass and the back element is a positive meniscus convex to the front of germanium. The refractive index of BS1 chalcogenide glass is 2.4914 (at 10 µm) and of IRG 100 is 2.6002. The temperature coefficient

of the refractive index is 70×10^{-6} for BS1 and 56×10^{-6} for IRG 100.

AGILTRON produces imaging lens system consisting of three chalcogenide lenses. These components are made of low cost and moldable chalcogenide glass that can accommodate both medium wavelength IR and long wavelength IR ranges. These unique features provide a best solution for a high resolution IR imaging with high signal intensity level and having the smallest size, lowest weight, and lowest cost.

We have developed a new simple procedure for producing the spherical and plano-convex microlenses based on chalcogenide glasses with emphasis on the As_2S_3 glass. Lenslets of micrometric size were fabricated by a procedure that allows for the production of a high amount of lenslets of various diameters and shape. The patented procedure [Patent deposed to OSIM, Romania April 2006] consists in passing the fine As_2S_3 powder through a gas flame that melts every piece of As_2S_3. The melted droplets fall onto a glass plate or in a recipient containing water at room temperature, and are cooled down rapidly. The droplets in the solid state can take the spherical form if the cooling is done in water and plano-convex shape if the cooling is made on glass plate.

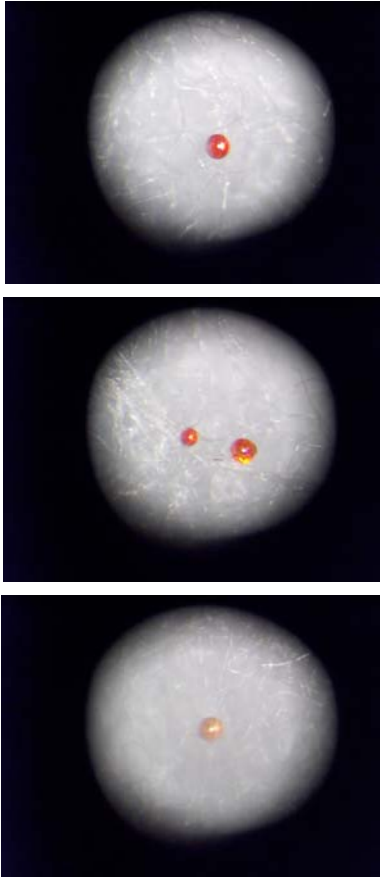Fig. 1 shows several lenslets obtained with the new, patented procedure.

Fig. 1. As₂S₃ lenslets of various sizes (~500 nm).

The lenslets and kinoform lenses are now developed in the direction of smaller focal lengthes with the aim to reach the nanometer length scales. As remarked by [99] the apperture of a single kinoform length is no longer limited by absorbation, and is instead limited by the critical angle. The limited aperture results in a limited optic resolution of $\lambda/\theta_c = \lambda\sqrt{2\delta}$. This λ/θ_c resolution limit is not a fundamental limit, and by using multiple kinoform lenses, this resolution limit can be exceeded. A compound kinoform lens of N lenses can produce a spot of size $\lambda/N\theta_c$. By increasing the number of lenses one could finally run into the true far field resolution limit, the wavelength of the focused photons. The limitation is due to the loss introduced by each lens leading to a signal-to-noise limit, not a resolution limit.

In the approach of Stein et al. [100] of the crossed – pair of cylindrical lenses, a fundamental limitation is the breakdown of the Fresnel approximation. In the Fresnel approximation spherical secondary wavelets are replaced by wavelets with parabolic wavefronts. After Goodman [101] the starting point for such an analysis is the Fresnel-Kirchhoff diffraction formula.

In the thin optic approximation one can write:

$$U(P_o) = \frac{1}{i\lambda} \int\int_{\xi\,\eta} U(P_1) \frac{\exp(ik\,r_{01})}{r_{01}} \cos\theta\,ds \qquad (3)$$

where $k = \frac{2\pi}{\lambda}$, and $r_{01} = \sqrt{z^2 + (x-\xi)^2 + (y-\eta)^2}$ with (x,y,z) in the image plane and (ξ, η) in the thin object plane. One can expand r_{01} in a series, and retaining terms to second order brings us to the Fresnel approximation for r_{01}:

$$r_{01} \approx z\left[1 + \frac{1}{2}\left(\frac{x-\xi}{z}\right)^2 + \frac{1}{2}\left(\frac{y-\eta}{z}\right)^2\right] \qquad (4)$$

showing the replacement of the spherical wave by a pair of orthogonal parabolic terms. Within the limits of validity of this approximation the crossed linear pair is separable, and each lens focuses independently of the other. By examining the higher order terms one can show that the limit of validity of this approximation is:

$$z^3 \geq \frac{\pi}{4\lambda}[(x-\xi)^2 + (y-\eta)^2]^2 \qquad (5)$$

Stein at al. [100] considered λ of order 0.1 nm and an aperture of 100 μm. They found that z, the focal length, is $\geq$ 1 cm, corresponding to a diffraction-limited focused spot size of the order of 10 nm. In other words, for 100 μm aperture lenses, it is expected a crossed lens pair to at least be able to focus down to 10 nm.

Fabrication of large format optical elements requires expensive and sophisticated equipment. The common way is to use multilayer structures got by thermal evaporation in vacuum. Electron beam evaporation is used as well but it has the advantage of relatively low energy of particles impinging on the substrate. Sputtering processes offer much higher energy. The ion beam deposition as well as chemical vapor deposition, have several advantages but

substrate size and location bring problems. The optical performance of optical elements including microlenses, got by any of the above noted methods is limited by the microstructure of the films and the quality of the substrate surface [101]. Holographic techniques are an alternative to such classical optical element fabrication. Holographic elements are periodic structures obtained with the help of interference patterns formed by mutually coherent beams. A hologram is a record of that pattern in a photosensitive material via modulation of the complex refractive index or the physical thickness of the material.

3. Chalcogenide lenslet arrays

Microlens arrays consist of an array of microscopic-sized lenses fabricated on a planar substrate and find applications in integrated optics, CCD cameras, faxes, image processing and optical computing [102].

Microlens arrays have been fabricated by a variety of techniques as e.g. (resin thermal reflow [103], laser ablation [105] or distributed index in planar technique [106]), but the most common and widely used technique is photolithography in which the photoresistive material that is deposited on a substrate is exposed through a mask by tailored light distribution. Usually 3D – structures having the form of the microlens arrays are generated in a photoresist material and then transferred by anisotropic etching into a robust material transparent in the spectral range for which the microlens arrays are intended. Microlens arrays have been produced in undoped As-S films [107] using the normal photo-induced solubility change that occurs in many chalcogenide compositions. After the array has been formed, a layer of Ag was evaporated over it and exposed to UV light. By photodoping Ag into the glass in this way it was possible to increase its refractive index (from 2.5 to 2.9) and, hence alter the focal length of the microlenses (from 34 μm to 27 μm).
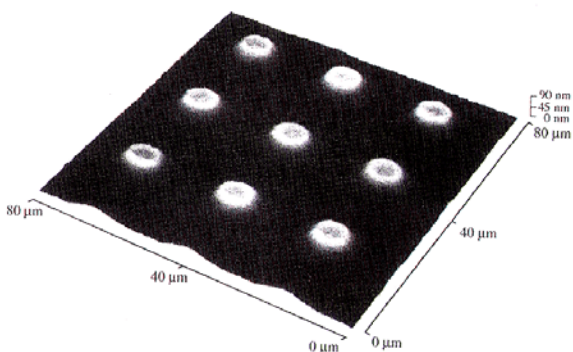

Fig. 2. Microlens array fabricated by Ag photodissolution into an AsS$_2$ film. The surface relief is 90 nm high and is due to the volume expansion that accompanies photodoping. (Atomic force microscope image). (after: P. J. S. Ewen in Photoinduced Metastability in Amorphous Semiconductors, Wiley - VCH, Ed. A. V. Kolobov, 2003, Ch. 22, p. 375).

A microlens array can also be fabricated directly using the metal photodissolution effect. One possible method has been demonstrated by Wagner et al. [108] who produced a 24 × 24 array of 20 μm diameter lenses by photodissolving silver into As$_{33}$S$_{67}$. In this technique, lens-shaped relief is produced by the volume expansion that accompanies the photodissolution. Fig. 2 shows an image of a part of microlens array as seen in the atomic force microscope.

Lyubin et al. [109] proposed a new technology for the fabrication of microlens arrays, which eliminates the necessity of anisotropic etching. This technology is based on the use of chalcogenide glasses that are simultaneously effective photoresists and very good IR optical materials. The method proposed is essentially the direct one-step formation of a 3-D microlens array using the dependence of the etching rate on the illumination intensity of chalcogenide photoresists. Cylindrical IR microlens arrays were fabricated with As-S and As-Se photoresists. The following parameters are typical for microlens arrays made with As-S photoresist of 0.35 μm thickness: diameter of single lens is 12 μm, pitch is 12 μm, focal length (for L = 500 nm) is 34 μm, array size is 40 × 40 microlenses, fill factor is 97 %. In spite of many advantages, these microlens arrays also have some drawbacks. The maximum sag in such microlenses is limited to 1.3 μm and the focal length can be made much shorter if the sag is increased. The reason for the sag limitation is the photodarkening of the chalcogenide film upon exposure. The photodarkening effect does not present a problem when low sag values are required below 1.0 – 1.3 μm, but due to this effect there is a self-limitation process that limits the thickness that can be exposed. Another drawback is a not very good shape of the convex surface formed in the direct one-step formation of microlenses. In order to overcome these drawbacks one can use the thermal reflow method that avoids exposure problems and leads to improvement of the shape of the lenses. Using a binary mask containing holes and slits, the islands of 3D binary shape can be formed that can be then transformed to 3D plano-convex microlenses. This is done by heating the material close to the melting point causing reflow and formation of the desired 3D shape. In a later research an AsSeI$_{0.1}$ photoresist (~820 nm) have been used due to its low melting temperature. A contact binary photomask containing slits for cylindrical microlenses was used. Samples were exposed using a 200 W halogen lamp and then developed in the negative photoresist mode in a monoethanolamine developer for 25 seconds in order to achieve 3D binary shapes. The thermal reflow method used here consisted of step heating of the binary shapes at the intervals of 10 °C from 170 °C to 240 °C for 5 minutes at each interval. The microlenses obtained by the thermal reflow method had a very good convex surface.

IR microlens arrays of big size have been fabricated using the thermal reflow technique due to development of a new very effective selected developer on the base of isomylamine. Spherical microlens arrays with the diameter of a single lens of 48.7 μm have been fabricated. Cylindrical microlens arrays with the length of a single lens of 2500 μm and width of 50.4 μm was also produced.

Figs. 3 and 4 show examples of spherical and cylindrical arrays of microlenses produced by the reflow method, described above.
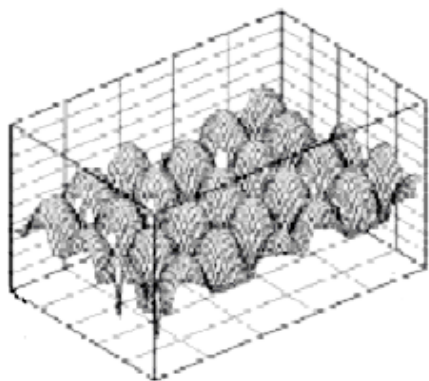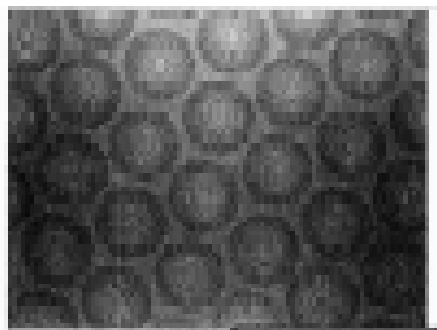
Fig. 3. Spherical microlens array based on As$_2$S$_3$ (after: N. P. Eisenberg et al. J. Optoelectron. Adv. Mater. 4(2), 405 (2002)).
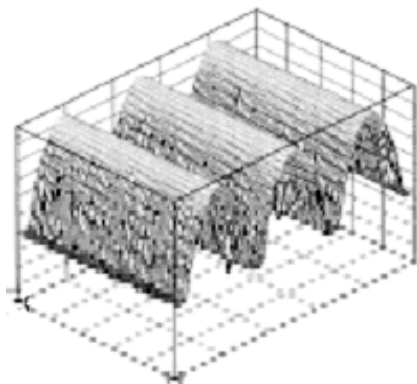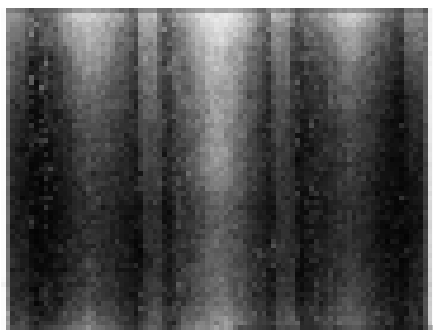

Fig. 4. Cylindrical microlens array based on As$_2$S$_3$ (after: N. P. Eisenberg et al. J. Optoelectron. Adv. Mater. 4(2), 405 (2002)).

In order to fabricate the microlens arrays, a 5 μm As$_2$S$_3$ photoresist was deposited by vacuum thermal evaporation onto an oxide glass substrate. There were used contact binary photomasks containing a set of transparent circles on a non-transparent background for spherical microlenses. The films were exposed to the light of a 1000 W xenon lamp and there after developed in the negative photoresist made in the isomylamine based developer for 8 minutes. This process resulted in a negative image as As$_2$S$_3$ islands of 3D shape. The thermal reflow procedure consisted of heating at 290 $^{\circ}$C for 5 min. Plano-convex refractive spherical and cylindrical microlenses were obtained.

4. Conclusions

The state of art of kinoform optical elements, lenses and microlenses based on chalcogenide materials is presented. The last achievements in the preparation of chalcogenide microlens arrays are discussed.

Acknowledgement

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