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Direct laser writing of two-dimensional photonic structures in amorphous As_2S_3 thin films

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A method for building photonic structures in As_2S_3 amorphous chalcogenide films has been developed. 2-D photonic configurations characterized by a regular assembly of rods (triangular lattice) or gratings with traces of micrometer period have been obtained. A femto-second laser has been used in photonic crystal registration. The simulation performed on these photonic configuration shows that the obtained chalcogenide structures could be efficient for operation in the infrared range around several micrometers wavelength.

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Keywords: Photonic crystal; Chalcogenide glass; As_2S_3; Thin film

1. Introduction

On chip integration of optical components promises to greatly enhance speed and reduce costs in optical communications. The so-called photonic crystals represent one of the most promising platforms for on-chip integration as they can combine optical waveguides, resonators, modulators or lasers [1]. The concept of photonic crystal and photonic band gap materials were introduced by Yablonovich [2] and John [3] as early as 1987. Nevertheless, up to day, the fabrication of not only 3-D but also 2-D photonic crystals is still challenging.

The photonic crystals or photonic structures based on amorphous chalcogenides are highly sensitive to fabrication imperfections [4], and therefore, it is important to control the defects of such lattices. Simple etching and interference lithography [5] are important techniques for revealing and correcting the photonic structures. A review on the advances in chalcogenide glass photonics is given in our review [6].

The tunable Ti-sapphire laser source is used for chalcogenide glass processing. The structures can be easily grown based on dynamical optical effects and two-photon absorption. Surface etching produces materials ablation and allows to create diffractive optical elements, quantum dots, etc… leading the way forward to evanescent wave couplers and sensors, as well as photonic configurations. As_2S_3 can be combined with semiconducting photonic crystals to create nanophotonic devices whose optical properties can be independently tuned on the same chip [7]. The development of optical devices with similar functionality to that which the transistor provides in electronics is a “holy grail” in photonics. A photonic transistor would allow the control of high-speed optical signals by light. This would simplify and lower the cost of future optical communication networks. The challenge is to fabricate devices that operate at low optical powers and at speeds of several tens of GHz.

With the purpose to inscribe in an easy way a photonic configuration and avoid the major defect occurring in a lot of materials we used a chalcogenide glass of composition As_2S_3. The material ensures the transparency in red and infrared regions of the electromagnetic spectrum (the absorption edge is situated around 0.53 μm wavelength). Moreover, the index of refraction is ~2.45, and thus the permittivity contrast glass-air of minimum 1.9 will be favourable to open the full photonic band gap. 2D photonic lattices have been inscribed with high accuracy and low amount of defects. A simulation experiment has been carried out before laser recording. The nanometer morphology, has been reached with great accuracy.

2. Experimental

The amorphous photonic materials, from the class of chalcogenide glass are excellent media for bandgap engineering by controlling the size and perfection of the unit cell configuration. Firstly, bulk As_2S_3 glasses were prepared by heating the powder of the compomemts in quartz ampoules above the melting temperature, followed by quenching in water.

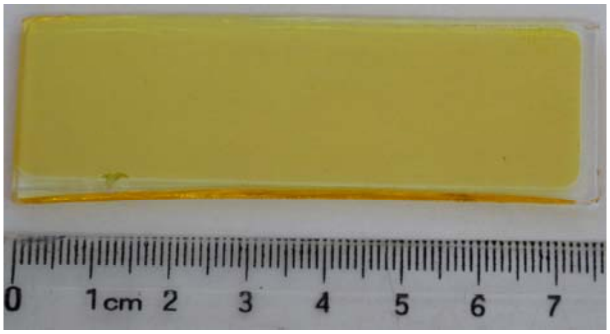

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Fig. 1 Thin As_2S_3 film deposited on glass substrate.

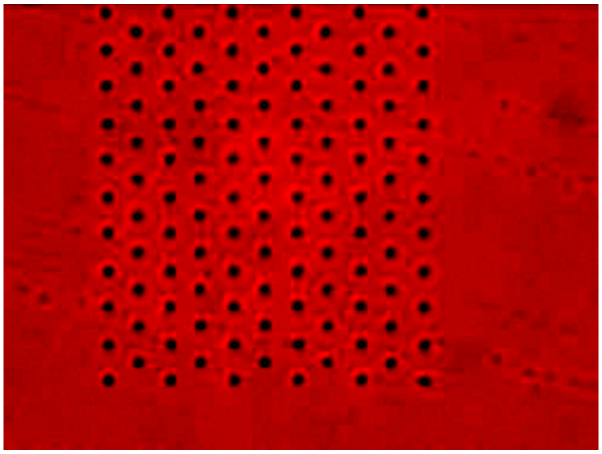


Fig. 3 Triangular lattice with rods in the As_2S_3 films

Thin amorphous films of ~ 1-2 micrometers thickness were deposited on optical glass by vacuum evaporation technique. A VUP 5 apparatus has been used for the deposition of thin films. Fig. 1 shows the image of an optical glass lamella covered by fresh evaporated films. The yellow color of the deposited film speaks in favor of the stoichiometric composition of arsenic trisulphide, as present in the target used for eposition. The crystallographic state of the As_2S_3 films has been checked by X-ray diffraction (Fig. 2). The X-ray diffraction pattern taken with CuK_α radiation does not evidence narrow diffraction peaks. The amorphous pattern shows a narrow "first sharp diffraction peak" around 8^o (θ) with the parameters (broadening and angular position on the θ scale) characteristic to arsenic trisulphide glass.

The cavity configuration in thin films has been fabricated by using a femtosecond Ti: Sapphire laser with the following working parameters:

- Power: 50 mW max., adjustable
- Pulse width: τ=15 fs enlarged to 50 fs, with new optical components installed
- Repetition rate: F=80 MHz, period T=1/F = 12.5 ns
- Wavelength: λ=775 nm
- Objective: ×100, NA=0.5, spot ~1.9 μm
- The calculated peak power: $P = P_m \cdot T/ \tau = 50\times12.5/50 \sim 10$ W,

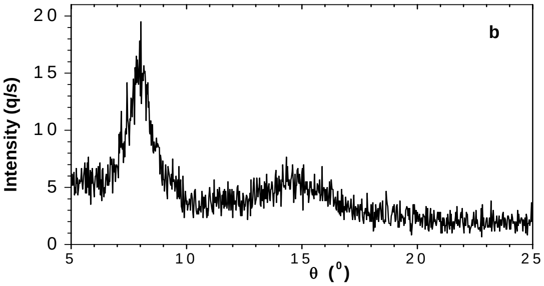


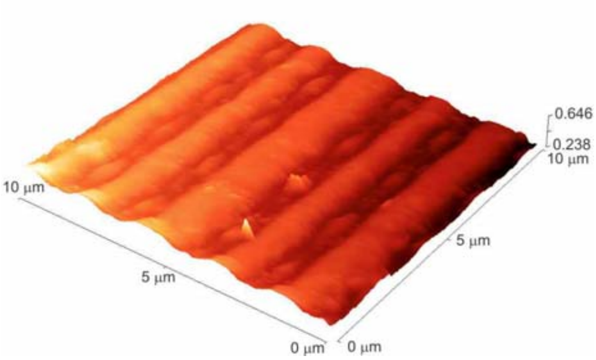
Fig. 2 X-ray diffraction diagram of the thin amorphous film of As_2S_3.

The As_2S_3 films with thickness 2 μm were prepared by at a vacuum of $\sim10^{-6}$ Torr on polished glass substrates and on silicon wafers. The laser wavelength 775 nm is choosed so that the linear fundamental absorption be negligible. However, short and high intensity femtosecond laser pulses undergo multiphoton absorption, especially two-photon absorption for this chalcogenide compound having band gap absorption near 530-540 nm wavelengths. The use of two photon absorption mechanism, instead of fundamental absorption one, is essential for the design of 3D photonic structures due to the fact that physical modifications in material appear locally and only when the exciting light exceeds certain intensity value. Thus, by using a laser beam it is possible to inscribe very accurate morphologies on amorphous chalcogenide films and to minimize the defects, which are inherent during the writing process.

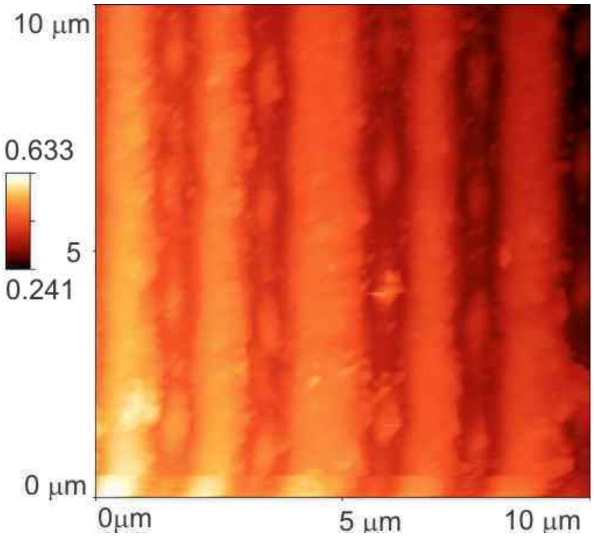
The substrate with the chalcogenide film was placed on mechanical XYZ scanning system with 15-20 nm positioning resolution. Computer software was developed in order to obtain the desired pattern. For example, a triangular configuration (Fig. 3) with lattice constant 5 micrometers has been inscribed in the As_2S_3 thin amorphous film.

The profile is obtained by developing the imprinted patterns in a selective organic base diethilene-triamine etchant. Washing in ammonia solutions with the concentration of 0.1 mol, as described in [8], has been done. The etching time was 30 s. The use of inorganic KOH for etching produces intense light scattering due to high level of the surface roughness obtained, as observed in atomic force microscopy (AFM) investigations. Several surface profiles have been obtained by etching. The profiles include photonic crystal configurations of different lattice constant and depth.

In the following pictures we show several AFM images (Fig. 4-6) where laser traces are visible, as well as point-like configurations (short columns) having the diameters in the micrometer range of values. It is remarkable the presence of ablated materials inside the traces from Fig. 4, as well as the particular profile of the small columns from Fig. 8 and 9.

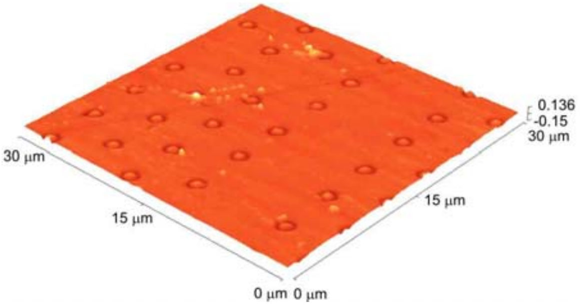


a) 3D picture

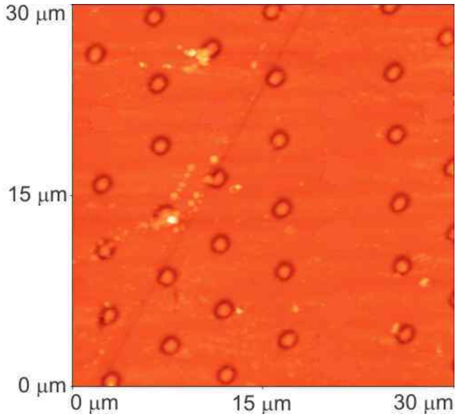


b) 2D picture

Fig. 4. Diffraction grating with a 2.0 µm period and 0.44 µm profile height Imprint of grating is done by scanning the focused light on the film. 10 × 10 µm AFM image. The refractive indices contrast ratio is 2.45:1 that allows the achievement of 2D and 3D photonic structures with total band gap open.



a) 3D AFM picture



b) 2D AFM picture

Fig. 5. Print of point by point periodical structure. Period is 5 µm, maximal height is 0.1 µm. Although the calculated spot diameter of the laser radiation is 1.9 µm, the diameter of an element is only 0.9 µm because lower exposure energies were used. This denotes that the exposure emerges at energies exceeding a certain level specific to non-linear processes (bi-photonic absorption in our case).

The controlled band gap modification can be introduced by creating linear defects (e.g. missing of one column and 1 row). These defects could be seen in the structure presented in Fig.5.

Of great interest is the investigation of the role played by the defects in the photonic structure. Fig. 6 and 7 shows typical defects produced accidentally during the inscription of the triangular and square photonic lattice of holes into glassy film. By controlling the defects, special 2-D configuration of points with different band gap opening in comparison with the band gap of the triangular lattice of rods can be created.

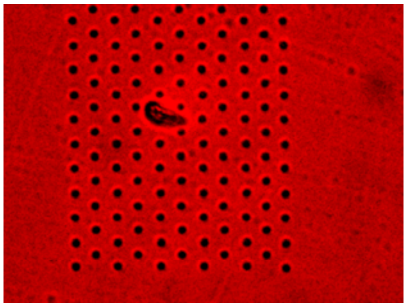


Fig. 6 Triangular 2D lattice

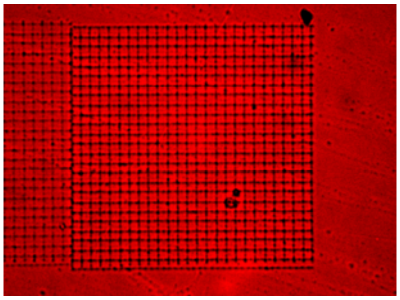
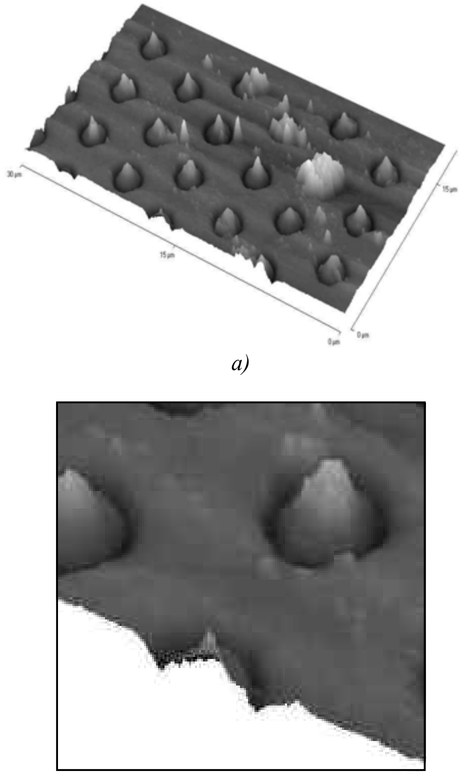


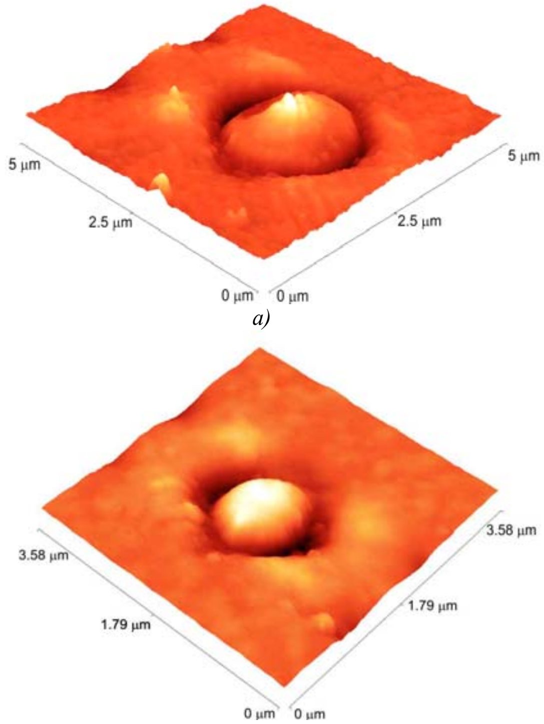
Fig. 7 Quadratic 2D lattice.



a)

b)

Fig. 8 Print point by point of a periodical structure. The period is 5 µm and the diameter of an element is 1.5 µm. (30 × 30 µm AFM image). a) 3D column profile in triangular symmetry realized by etching. b) Detail of a column.



a)

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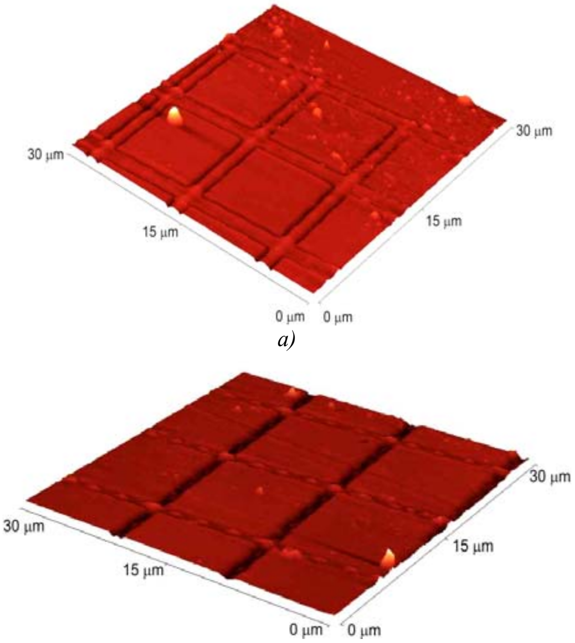
Fig. 9. a) One point profile obtained with higher laser energy. b) The same obtained with lower energy. Dots with dimensions smaller than laser spot are observed.



a)

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Fig. 10. Two examples of structures obtained by scanning with laser beam. a) Structure obtained at a higher energy level, b) the structure on the sample exposed at a lower energy level.

In Fig.8b you can clearly see a deep valley created around the central column. The nature of the valley is not yet clear. It occurs both for high and low levels of energy (Fig. 9 a,b). A possible cause may be related to bi-photonic absorption mechanism. Anyhow, this phenomenon is not produced by a non-uniformity of the laser beam.

The investigations of writing laser intensity distribution demonstrate good Gaussian shape as can be seen in the next picture (Fig. 11).

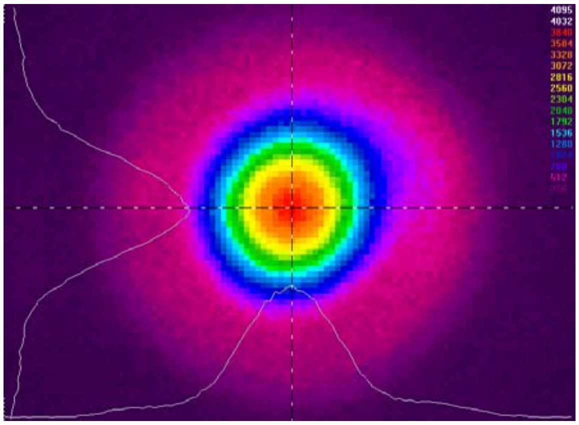

Fig. 11. Far field profile of writing laser beam shows Gaussian shape.

The Gaussian shape of the laser beam cross-section can explain the round shape of the micrometer dots inscribed on the surface of the amorphous chalcogenide film As_2S_3 and evidenced by etching.

The dimension of the rods or holes patterned into special configurations control the photonic band gaps as a function of lattice constants. Numerical calculations must be done in order to characterize the bandgaps.

3. Simulation of the photonic bandgap.

Numerical simulations were performed using the MPB-MEEP codes developed by Joannopoulos et al. [9]. More precisely, the tri-holes and tri-rods code too was run to find the band-gaps for the dielectric constant $\varepsilon = n^2$ belonging to the interval 6.0-6.5. This interval corresponds to the refractive index of material equal to 2.45-2.55 in the near IR region. Permanent refractive index modification in As_2S_3 can be done by CW green laser irradiation. Results of simulations are shown in Fig. 12 and Table 1.

Fig. 13 shows the scheme of the photonic band gap in the amotphous As_2S_3 film.

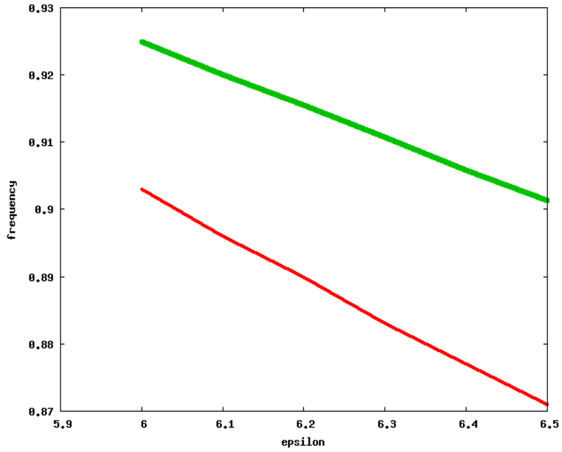

Fig. 12. Band-gap edges in ωa/2πc units vs. dielectric constant ε. Upper edge – green; lower edge – red.

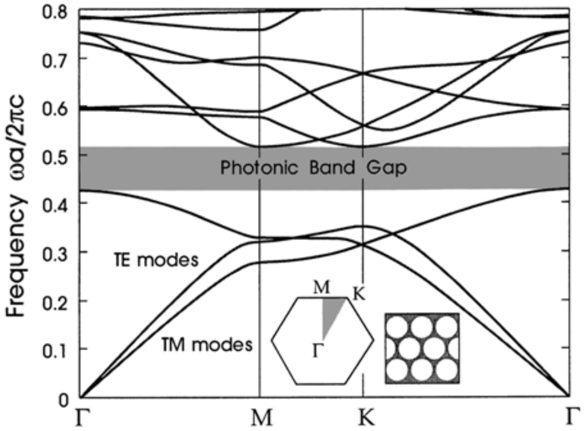

Fig. 13. 2D photonic gaps in As_2S_3 films.

Numerical simulations on tri–rods lattice were performed too, with the same lattice constant and the same dielectric constant. The radius r of the rods was varied in the interval 0.15 – 0.4 (fractions of period a) with an increment of 0.5. The corresponding band-gaps obtained are displayed in the Table 1.

Table 1 Photonic band gap in As_2S_3 films.

r (fraction of $a = 1\mu m$)	Band-Gap (Hz×10^{14})	Band-Gap (%)
0.15	3.18 – 3.24	1.7
0.20	2.82—3.03	7.4
0.25	2.72 –2.76	1.2
0.30	2.99 – 3.02	0.9
0.35	2.43 – 2.44	0.1
0.40	2.17 – 2.18	0.1

We observe a maximum of 7.4% in the band-gap for $r = 0.2$ fraction of lattice period.

4. Discussion

The currently developed devices can be optically tuned by using photoinduced changes in As_2S_3 glass, so post-tuning of the photonic components is possible. The illumination of chalcogenide based photonic devices with light having energy higher than band gap initiates a structural rearrangement in glass. Thus, the properties of the glass are significantly changed. As a consequence the hardness and the etching properties are modified and the chalcogenide becomes a negative photo-resists. Thus, strong relief appears on the glass (2-D configuration) and the photonic properties are imprinted after developing in a selected etchant.

It seems impossible to arrange a photonic crystal with both isolated spots and connected regions of dielectric material. The result is a compromise: we can imagine crystals with high-ε regions that are both practically isolated and linked by narrow veins.

Under suitable conditions the forward ejected material produces submicron and micron dots that are spread on the surface of the films. Thus a QD lattice could be prepared as a by-product of the photonic crystal technique if the surface of the film is carefully patterned.

In recent years the fabrication of photonic crystals using direct drawing by focused femtosecond laser beams, via two-photon absorption, received much attention. The main merit of the direct laser writing stems from its ability to produce almost any kind of three-dimensional architectures [10].

The direct laser writing is ideal for three-dimensional photonic crystals with intricate periodic configuration such as the diamond lattice [11], slanted pores [12] or spiral diamond structure [13]. All other known methods for writing proved to be unsuccessful.

In this paper we have demonstrated the possibility to inscribe not only photonic crystals but also various surface configurations at the micrometer and even submicrometric scale in a rather simple and stable chalcogenide glass: arsenic trisulphide.

Our future researches will be directed to the construction of two and three dimensional photonic crystals based on chalcogenide glasses with refractive indices higher than that of As_2S_3, as e.g. As_2S_3-As_2Se_3 $n \leq 2.6$ or AsSeTe ($n>3$). The absorption regions of the high-refractive-index glasses are shifted into the red edge of the visible spectrum.

The interference lithography seems to be a very useful method in developing many kinds of photonic crystals and devices integrated in optoelectronic circuits at the nanometer scale. If the structure of the arsenic sulfide corresponds to molecular As_4S_4 (Realgar) then the polymerization of the film during light irradiation will be maximized. Thus the etching will confer high contrast between the illuminated and non-illuminated zones and the photonic structure will be more clearly expressed.

5. Conclusions

A method has been developed for the production of photonic structures based on chalcogenide glassy (amorphous) films. 2-D photonic configurations characterized by a hexagonal assembly of rods (triangular lattice) or having gratings with traces of micrometer width have been obtained.

The waveguides and rods were created in As_2S_3 glassy films in a simple way with the help of a femtosecond laser and direct laser writing with submicron resolution and 20 nm positioning.

The procedure is simple, rapid and the borders of the lines and dots drawn on the chalcogenide films are very fine without ripples. The surface etching evidences smooth structures with only few defects or ejected material remained after the recording of the photonic structures. The remainders of the chalcogenide material can be further eliminated by washing in alkaline solutions.

Photonic structures can be formed at the surface of amorphous arsenic trisulphide films with great accuracy and resolution at the nanometric scale.

Acknowledgement

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