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Laser deposition of waveguiding Ti: sapphire and chalcogenide glass AsS films

J. Lanèok, M. Jelínek, L. Jastrabík, L. Soukup, J. Oswald and K. Jurek
Institute of Physics, Academy of Scinces of the Czech Republic,
Na Slovance 2, 182 21 Prague 8, Czech Republic

F. Flory
Laboratoire d' Optique des Surfaces et des Couches Minces, Ecole Nationale Supèriere de Physique de Marseille,
Dom. U. de St. Jèrome, 13397 Marseille, Cedex 20, France

M. Frumar
Dept. General and Inorganic Chemistry, Faculty of Chemical Technology, University of Pardubice,
53210 Pardubice, Czech Republic

Waveguiding Ti:sapphire on quartz and chalcogenide glass on fused silica substrates were prepared by pulse laser deposition. Ti: sapphire films were deposited at 500 ^oC from 0.48 wt% of Ti_2O_3. After annealing the high textured films were obtained, exhibiting attenuation 6-7 $dBcm^{-1}$ were obtained. Inhomogeneity of thickness and refractive index of deposited films were checked by m-line technique. Chalcogenide $As_{40}S_{60}$ films were deposited at an energy density range from 1.9 J/cm2 to 5.7 Jcm^2. It was found that the stoichiometry, optical density in visible spectral region, index of refraction and waveguiding properties of $As_{40}S_{60}$ films changed with deposition conditions.

*Keywords***:** laser deposition, thin films, Ti: sapphire, chalcogenide glass

1. Introduction

Passive and active planar waveguides are components of integrated optics and optoelectronics for generation and processing of visible and near infrared signals and development of a new generation of integrated optics technology in which sources, non-linear structures and detectors will be produced on a single substrate.

Recently coherent sources based on waveguiding Nd:GGG [1] and Ti:sapphire [2] layers were successfully created by a method of pulsed laser deposition (PLD). The PLD is now widely used for the study of various films for active and passive waveguiding layers [3].

The Ti:sapphire layers for the waveguiding laser were produced on sapphire substrates. On quartz the thinner layers would be sufficient to observe guided properties because of the higher difference in the index of refraction of active layer and substrate. But the attempts to fabricate layers on quartz substrate by a direct laser deposition were not fully successful because of the cracking of the quartz substrate with fast temperature changes [4]. This is the reason we tried to deposit Ti:sapphire on quartz at low temperature and then anneal the film to reach crystalline structure.

Chalcogenide glass films are a relatively new branch of waveguiding films. In recent years chalcogenide glass films such as Ga-La-S [5] and Ge-Sn-Se [6] were created by PLD.

We focused attention on $As_{40}S_{60}$ (AsS) films, which can be used for example for submicron photoresist, diffraction elements, optical waveguide, [7-8].The AsS films were created by PLD at various deposition conditions and the optical properties of films in relation to film composition were carefully studied.

2. Experiment

Films were deposited by KrF excimer laser ablation Lumonics PM 840 operating at 10 Hz with a 500 mJ per pulse.

For Ti:sapphire films a polycrystalline target with 0.48 wt% of Ti_2O_3 was used. The beam was focused on the target with an energy density ~ 3.7 Jcm^{-2}. The laser spot on the target (**x**,**y**)had on area 2.7x1.5 mm^2. The vacuum chamber was evacuated to a base pressure of 10^{-3} Pa. The quartz substrate had a (001) orientation. The substrate was fixed on the massive resistively heated substrate holder and placed 45 mm away from the target. Deposited films were post**-** annealed in the first step in an oven in an open air atmosphere at 800 ^oC for 1 hour. After analysis the films were again annealed at 1100 ^oC for 5 hours in a flowing oxygen atmosphere. To avoid film cracking the rise and the fall time of annealed temperature changes was very small (2 ^oC min^{-1}).

The structural properties of Ti:sapphire films on quartz were analysed by XRD by means of the standard Bragg-Brentano alignment employing a Cu rotating anode. The luminescence and waveguiding properties (index of refraction and attenuation) were also checked for deposition and annealed samples.

For the luminescence measurement an Ar^+ ion laser was used as excitation source was used. Waveguiding properties were measured by m**-**line technique by using a He**-**Ne laser (λ=632.8 nm). The refractive index and geometrical thickness of the films was measured on different places of the film.

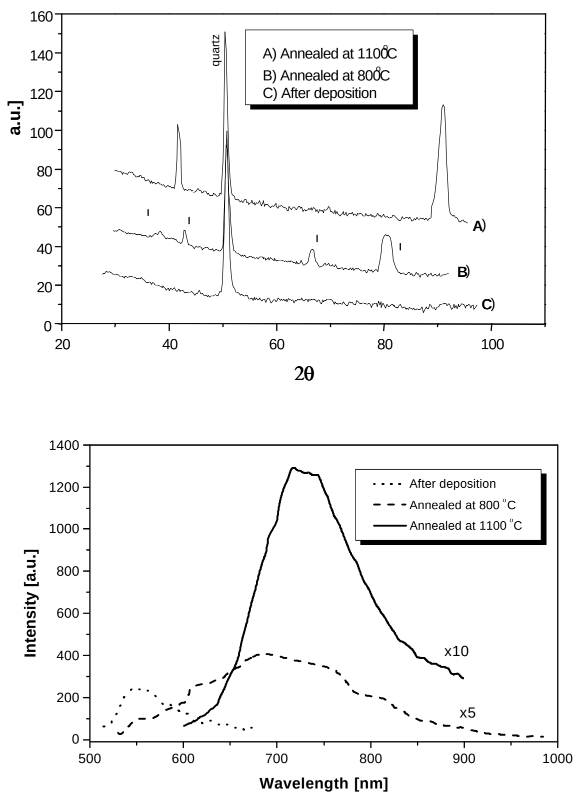

Fig. 1 XRD spectra (**a**) and luminescence (**b**) of as deposited and annealed Ti: sapphire layers created on quartz substrate

The experimental set-up consists of the He-Ne laser and prism in configuration similar to that presented in fact [9]. Attenuation of annealed films was determined at 632.8 nm from the measurement of scattered light coupled out of the guide along the propagating path [10].

The chalcogenide glass AsS films were created from $As_{40}S_{60}$ bulk substrate at room temperature. The energy density of KrF radiation on the target was changed from 1.9 Jcm^{-2} to 5.7 Jcm^{-2}. The chamber was evacuated to 10^{-2} Pa. The target was prepared by the melting of purity (5N) elements in evacuated quartz ampoules (650 °C-700 °C) for a duration of 12 hours. The distance betwen forget acid substrate was 40 min. Film stoichiometry was measured by WDX (Wavelength Dependence X- ray analysis) - i.e. electron superprobe JEOL JXA 733 with microanalyser KEVEX D). The absorption spectra in the visible region were measured by a Shimadzu UV 1601 spectrometer having a resolution of 2 nm. The index of refraction of the films was calculated from m-line measurements using the admittance method [11].

3. Results and discussion

Ti: sapphire films

The Ti:sapphire layers deposited at 500 °C were amorphous.. After annealing at 800 °C the films were polycrystalline and four peaks of Al_2O_3 in XRD spectra were observed:(1$\bar{2}$10), (1$\bar{2}$13), (2$\bar{3}$14) and (2$\bar{4}$20). After annealing at 1100 °C the film exhibited highly crystalline orientation. On the diffraction pattern only peaks from (0001) crystalline planes of sapphire are presented - see Fig.1a. From the width of the diffraction maximum (0006) and (00012) it is possible to predict the high level of stress, which is presented in the film due to the lattice mismatches between the sapphire and quartz. For this reason the films of thickness ~1 μm peeled spontaneously off after 1 week. Only films of thickness of about 0.5 μm were stable and were used for analysis.

Due to the dependence of luminescence of the Ti^{3+} ions on surrounding crystalline field, the luminescence spectra also changed along with crystalline changes of the films during the annealing process. The maximum of the luminescence spectra of amorphous and polycrystalline films moved to the blue part of the spectra - 550 nm (for amorphous film) and 690 nm (for annealed film at 800 °C). The luminescence of the monocrystalline film has its maximum at 730 nm, which corresponds to the luminescence spectra of the Ti:sapphire bulk [12]- see Fig.1b.

The refractive index of Ti: sapphire films also changes with thermal annealing. The refractive index of amorphous films after deposition was 1.6929, after annealing at 800 °C the refractive index decreased at 1.6905 that can be caused by a better oxidation of Ti inside the film. The index of refraction of the crystalline film increased to 1.7585, which is close to the value of the Ti:sapphire bulk. Attenuation of crystalline films was found to be 6-7 dB/cm (for a film with a thickness of 450 nm), but depends very much on the position of the tested He -Ne beam in the layer.

The experimental setup for thickness nonuniformity measurement is show in Fig.2. The (Δθ) shift of the m- line was observed with the motion of the laser spot (Δχ) oN the prism base. This effect can be caused by nonuniformity of the thickness and/or refractive index of the film. From the angle of synchronisms of the supported modes, the change of the refractive index and the thickness was obtained as a function of **x** (corresponding with laser spot orientation). From the calculations it follows that the refractive index of the film is constant and is equal to 1.7585±0.0005 along all to the film

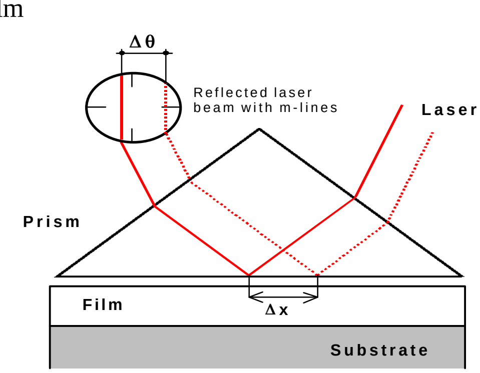

Fig. 2. The experimental setup for measurement of the thickness nonuniformity by using m-line technique

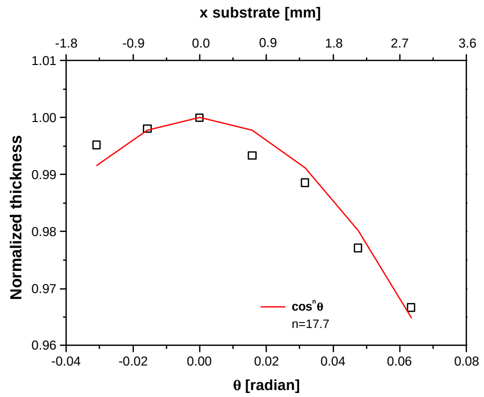

Fig. 3. the thichness nonuniformity of the Ti: saphire film. the maximum film thickness was 460 nm.

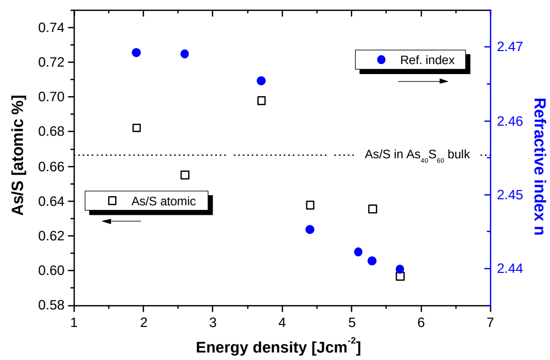

Fig. 4. Change of chalcogenide AsS glass film stiochiometry ( ) and film refractive index (•) with deposition energy density

. Due to this fact the motion of m-line was caused only by thickness nonuniformity. The measurement of the film thickness nonuniformity is shown in Fig.3. to obtain the thickness distribution a fit was performed to the function Cos ^{n} (θ), with (θ) the angle measured from the target normaly obtaining n=17.7±1.2. According to [13] the natural thickness changes of films introduced during the PLD growing procedure can lead to an increase of the confinement factor of the optical radiation and does not significantly affect the overlap of the pump and signal fields inside the active layers.

Chalcogenide AsS glass films

It was found that the parameters of the AsS films are significantly changed with deposition conditions. The change of film stoichiometry with deposition energy density is show in Fig.4. We see that the film is slightly sulphur deficient for low energy density (1.9-.3.8 Jcm^{-2}), while the film deposited at high energy density (more than 4 Jcm^{-2}) is slightly arsenic deficient compared with the expected composition of $As_{40}S_{60}$. The oxygen content inside the film was also measured, the concentration of oxygen was very small and varied between 0.1 and 0.6 atomic percent.

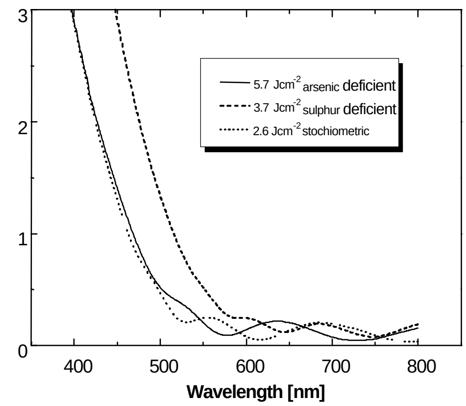

Fig.5. Absorption spectraof AsS films deposited at various laser energy density

The film, with a composition desert to that of the bulk was deposited at an energy density 2.6 Jcm$^{-2.}$ and has stochiometry $As_{39.6}S_{60.3}$.

The refractive index of the films deposited at lower energy density (1.9-4 Jcm^{-2}) is higher, around 0.03, than the refractive index of the films deposited at a higher energy density (more than 4 Jcm^{-2})-see Fig.4. The mode spectrum of the films was also checked by means of the m-line technique. Depending on the film thickness, the 2 -4 TE guided modes were observed on the films.

Fig.5. shows the dependence of the optical density ($\log_{10}$ 1/T, T-transmision) on the wavelength. The periodic modulation observed is an etalon effect due to light interfering from the air/film and film/substrate reflections.

The stochiometric film and slightly arsenic deficient films have cut-off wavelength around 400 nm, while the sulphur deficient film has absorption edge shift to the red part with cut-off wavelength around 450 nm.

4. Conclusion

Monocryatalline Ti:sapphire films with crystallographic orientation (0001) were obtained after thermal annealing of amorphous films deposited by PLD on quartz substrate. The changes of the optical properties of the films such as luminescence and refractive index with the crystalline structure were measured. Compared with amorphous films the luminescence intensity of crystalline films rapidly increases and shifts to the red part of the spectra, with a maximum at 730 nm. The refractive index also increased from 1.6929 (amorphous film) to 1.7585 (crystalline film). The attenuation of Ti:sapphire films grown on quartz substrates at lower temperatures, with subsequent annealing, was found to be around 6 - 7 dB/cm. Similar results were obtained in the case of direct laser deposition on quartz at higher temperatures, but for lower Ti_2O_3 concentration in the film [4]. The thickness profile was successfully measured by m-line technique and exponent *n* in the curve profile cos^{n}(θ); n=17.7 was determine.

The $As_{40}S_{60}$ chalcogenide glass thin waveguides were successfully deposited at fused silica substrate. The dependence of film stochiometry on laser energy density was studied. The stochiometric film was deposited at 2.6 Jcm^{-2}. The refractive index of chalcogenide films was measured at 632.8 nm wavelength by m-line technique. By this method the waveguiding mode spectrum was observed and the 4 TE modes were observed. The absorption spectra in the visible region were measured for films with different compositions and the shift of the absorption edge to the red part of the spectrum for sulphur deficient film compared to the stochiometric film was observed.

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