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PREPARATION AND PROPERTIES OF LANGASITE AND YAG AMORPHOUS FILMS

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Amorphous thin films of langasite and YAG have been prepared from crystals targets by pulsed laser deposition, at room temperature, in vacuum, on silicon wafers. Bulges of micrometer size are formed on the film surface. Larger size bulges (10-50 micrometers in diameter) are characteristic to the annealed langasite films. Annealing at high temperatures leads to the crystallization of the films. The annealed langasite films (850 ^{o}C) are polycrystalline and partially oriented with the plane (001) parallel to the surface of the silicon wafer. The bulges break easily in the heat treated films. Their empty structure was remarked on the electron microscope images. The YAG films annealed at 1100^{o} C shows a crystalline YAG phase depleted in Y and traces of Y_2O_3. An interesting feature is the presence of silver particles spread along the macrodefects (fracture lines) of the film.

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1. Introduction

The interest in the lanthanum gallo-silicate, $La_3Ga_5SiO_{14}$, or langasite, rose tremendously due to its piezoelectric properties, analogous to those of quartz. Dubovik et al. [1] have shown that the functional parameters of the piezoresistive elements are better than those of quartz: the resonance interval is 2-3 times greater, the inductance and dynamical resistance for the bulk acoustic waves (BAW) are 6-18 times and 2-6 times lower, respectively. The electro-mechanical coupling coefficient is 2-3 times higher and the losses for the surface acoustic waves (SAW) are two times lower. Moreover, the size of the piezoresistive elements is reduced by 20-30 %.

Sacharov et al. [2] produced monolithic filters for GSM, based on langasite. The most promising field of such filters is that of the devices with surface acoustic waves (SAW) and bulk acoustic waves (BAW).

The outstanding dielectric properties of langasite makes it an important material in microelectronics for ultrathin insulators coatings.

The amorphous films based on langasite have been not prepared up today, after our knowledge.

YAG (yttrium aluminium garnet, $Y_3Al_5O_{12}$) crystals are important as a laser material. The doped YAG crystals doped by Er are luminescent. Few papers on the production of amorphous films of YAG have been reported up to day [3]. No data on the physical properties of such films are available. Pulsed laser deposition is a new method for the preparation of thin films.

This paper reports the preparation of thin amorphous films of langasite and YAG by pulsed laser deposition and the structural properties of fresh and annealed films.

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2. Experimental

In the last years various films were deposited by the method of pulsed laser deposition, and investigated [4-8].

Thin langasite and YAG films were deposited by pulsed laser deposition (PLD) using a KrF^* excimer laser (248 nm wavelength of the emitted UV pulse, pulse duration < 10 ns, 1Hz repetition rate, and maximum output energy of 85 mJ/pulse). The pulse was focused on the target through an MgF_2 cylindrical lens (focal length: 30 cm) argon coated. The incidence angle to the target was 45^o. The laser spot was set within 4.2 and 6.4 mm^2 and the incident fluence varied roughly in the range 0.8- 1.6 J/cm^2.

The target material were platelets of $15\times15\times2$ mm^3 cut from bulk langasite and YAG crystals grown by Czochralski method in our laboratory using high purity elements. The target holder was placed in a stainless vacuum chamber, which was subsequently evacuated down to $1\text{-}7 \times 10^{-4}$ Pa. The target was rotated with the frequency of 0.4 rot/min during PLD process. The substrate for film deposition was a [100] oriented silicon wafer mounted on a molybdenum heating block, parallel to the target surface and placed at a distance of 2-5 cm. The number of pulses applied for making one film was 30 000. The deposition was made in three successive stages.

From the ellipsometric mesurements we have found a thickness of 1.9 μm for the langasite film. This means a deposition rate of 0.63 Å/ pulse. A refractive index of 1.84 at 700 nm was determined. In the case of YAG film the thickness is considerably lower but the refractive index is similar to that of the langasite film.

The structure of the films has been determined by X ray diffraction and scanning electron microscopy. A TUR M62 diffractometer provided with copper target tube was used. An Amray 1830 I scanning microscope was used for the microscopy studies.

3. Results

3.1 X-ray diffraction

The films were investigated firstly by X-ray diffraction.

The fresh films of both langasite and YAG are amorphous (Fig. 1 a, b). An intense first maximum and a large, weak second maximum are the most important characteristics of the X-ray patterns.

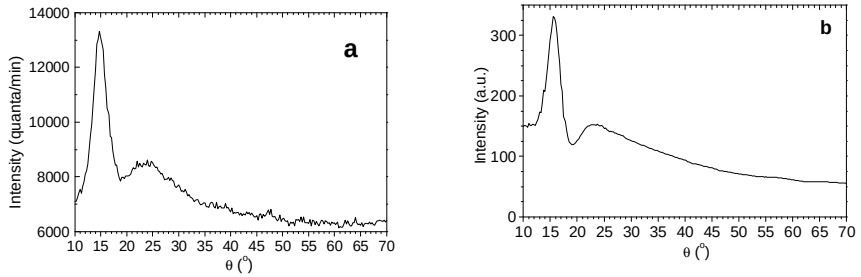


Fig. 1.a. The X-ray diffraction diagram of langasite film prepared by PLD. b. The X-ray diffraction pattern of the YAG film prepared by PLD.

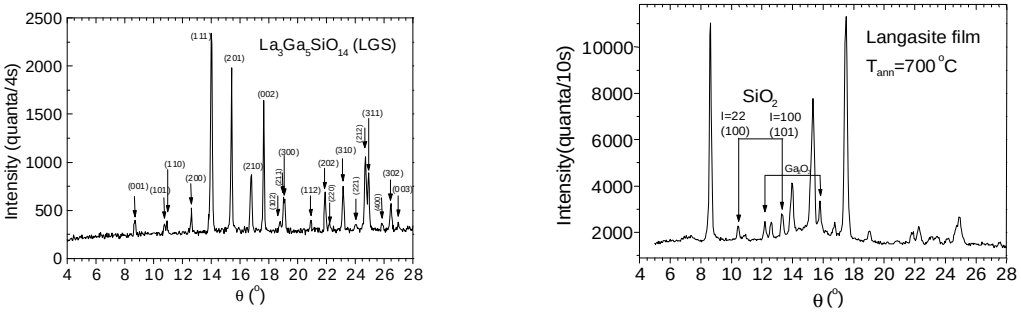
The first intense peak of the YAG film is significantly shifted towards large angles if compared to the intense peak of langasite films.

Two films of langasite were prepared. One film was annealed at 700 ^oC for 2 hours and the other was annealed at 850 ^oC for 1 hours. The amorphous film YAG was annealed at 1100 ^oC for 1 hour.

Fig. 2a illustrates the X-ray pattern of a langasite powder prepared from a small single crystal of langasite. The X-ray difraction diagrams recorded on annealed films of langasite and YAG are shown in Figs. 2b, 3 and 4.

Narrow peaks observed on the diagram of langasite and no amorphous background reveal the full crystallization of the film (Fig. 2b). The major part of the peaks can be ascribed to langasite. The high intensities of the (001) and (002) lines speak in favour of a strong preferential orientation of the langasite crystallites on the wafer substrate. In the case of annealed film the minor phase, evidenced as small peaks on the diagram,can be ascibedto La_2O_3 and SiO_2, the oxides resulted from the decomposition of langasite. The third oxide, Ga_2O_3, cannot be seen due probably to its vaporization during annealing.

High temperature treatment of langasite film induces the formation of highly oriented crystalline langasite phase (Fig. 3). Broad peaks indicate small size crystallites.



a b

Fig. 2. a. The X-ray diffraction diagram of a powder obtained from a single crystal of langasite. b. The X-ray diffraction diagram of the langasite film annealed 2h at 700 °C.

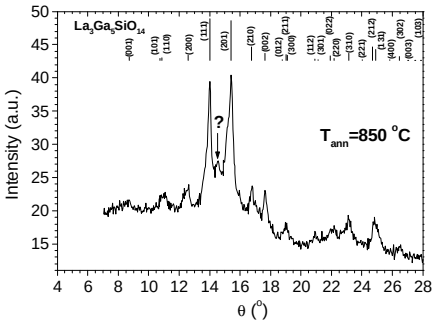


Fig. 3. The X-ray diffraction diagram of the langasite film annealed 1h at 850 °C.

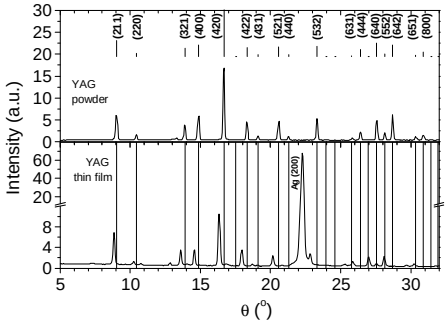
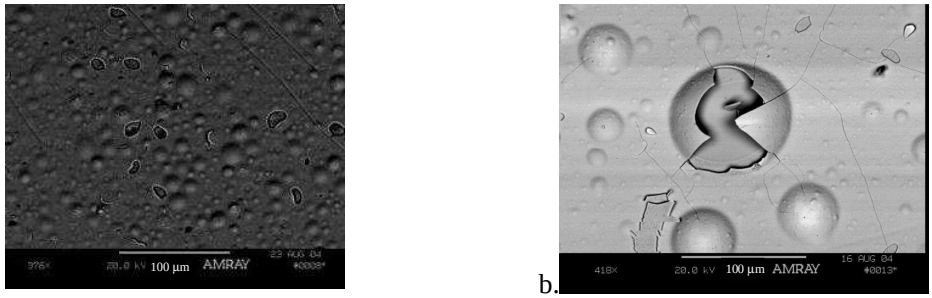


Fig. 4. The X-ray diffraction pattern of the YAG film annealed at 1100 °C for 1 hour.

Fig. 4 shows the X-ray diffraction pattern of YAG powder and of the YAG film annealed at 1100° C for 1 hour. The annealed film is crystalline. High temperature treatement of YAG film induces the formation of crystalline phase of YAG, somewhat depleted in ytrium. The sub-stoichiometry of YAG films was proved by the lattice parameter, a, of the YAG-type phase. In the stoichiometric phase a=12.016 Å, while in the non-stoichiometric phase (our films) a= 12.253 Å (calculated from the peak positions). A minute amount of crystalline yttrium oxide was detected in films.

3.2 Scanning electron microscopy.

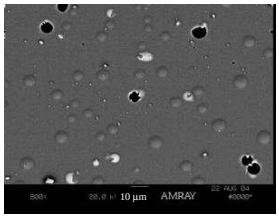
The scanning electron microscopy shows interesting features of the pulsed laser deposited films of langasite and YAG.



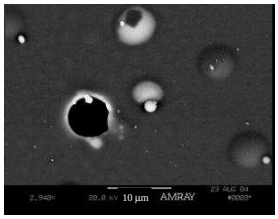
a. b.

Fig. 5. Scanning electron microscopy on langasite films annealed at low temperature (500 °C) (a) and at 850 °C (b).

Fig. 5a shows the morphology of the langasite film annealed at 500 ^{o}C. This treatment maintains the film in the amorphous state. Bulge-like defects with the diameter of ~5-20 μm are visible on the picture. By annealing the film at 850 ^{o}C, crystallization takes place. The morphology of the films is preserved but the bulges are evidenced as empty microzones and sometimes the empty bulges break and the substrate can be viewed (see Fig. 5b). The biggest bulge has the diameter of 60 μm.

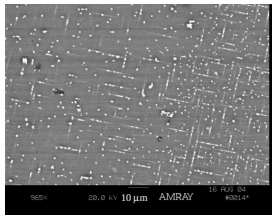


a

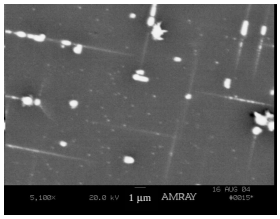


b

Fig. 6. a,b shows the morphology of the surface of the YAG film after annealing at 500 ^{o}C.



a



b

Fig. 7. a, b The morphology of the surface of the YAG film after annealing at 1100 ^{o}C.

Fig. 6 a,b show the morphology of the YAG film, still amorphous after annealing at 500 ^{o}C. Bulges with diameters of 5-8 μm are visible on the micrographs.

The YAG film annealed at 1100 ^{o}C exhibits an interesting feature (see Fig. 7 a,b). The surface is decorated in a geometrical manner by small dots, which have been demonstrated in the electron microscope (fluorescence device) to be made of silver. Silver is crystalline and oriented with the crystallographic plane (200) parallel to the substrate. The origin of silver seems to be the silver paste used on the back side of the deposited sample (silicon wafer) that, during annealing, have been evaporated and partially transported on the YAG film surface. The decoration follows, probably, the microcrack lines on the crystallized surface of the film. Earlier, we observed similar decoration on heat treated alumina films.

4. Conclusions

Thin amorphous films of langasite ($La_3Ga_5SiO_{14}$) and YAG ($Y_3Al_5O_{12}$) have been successfully deposited by pulsed laser deposition (PLD). The film morphology is characterized by homogeneous regions with embedded bulges. After thermal annealing, polycrystalline structures are formed. Langasite phase and YAG phase depleted in yttrium have been revealed. Annealing at 850 ^{o}C gives rise to small size crystallites of langasite, due probably to rapid firing (got by putting the sample directly at this temperature) as opposite to the case of annealing at 700 ^{o}C, where the heating of the furnace was made with the samples introduced in it. The advantage of the rapid firing is the prevention of phase separation. The bulges observed in the samples have diameters of 10-50 μm for langasite films and 5-8 μm for YAG films. During annealling the diameters of the bulges increase, and some bulges break, showing an internal empty structure.

The silver from the rear part of the wafer substrate of the YAG films is evaporated during high temperature annealing and redeposited on the film surface, as small white particles, which decorate the defects, and put in evidence the fine fracture lines in the film.

Acknowledgements

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