Crystallization Double-Layer Magneto-Active Films for Magnetophotonics

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Abstract. Magneto-optics, magnetophotonics and magnetoplasmonics stay at the edge of scientific interests last years due to their unique features to manage the light and electromagnet field. Bi-substituted iron garnet (Bi:IG) is one of most promising magneto-optical material for these applications in order to its high efficiency in visible and infrared spectra. The possibility to integrate Bi:IG films to silicon semiconductor process leads to creation nanoscale high performance magneto-optical devices. Bi:IG structures of different composition might be deposited by vacuum deposition on different substrates. The investigation of crystallization process of Bi:IG double-layer films at a different process parameter on gadolinium gallium garnet and fused quartz substrates allowing to determine dependences and suggestions for integration Bi:IG to semiconductor process or multicomponent optical nanostructures.

1. Introduction
Today scientific community actively develop the idea of design magneto-optic and optomagnonic structures that combine the optical effects of transmission / localization of optical waves and the dynamics of magnetization at femto- and picosecond times [1-6]. Rare-earth garnets and iron garnets have crystal structure which allows to achieve advanced features of magnetic system. Nowadays more and more efforts are paid to implement bismuth and cerium to the garnet lattice. It may significantly increase the magneto-optic (MO) performance. Possibility of garnet materials to be deposited on the crystals without garnet structure opens the door to semiconductor process. The MO films’ integration to silicon semiconductor process is not easy to implement but important for fabricating new nanoscale and high-performance devices [7]. Real synthesis of magneto-active structures meets a lot of difficulties in the way of using different garnet composition as a part of nanoscale one-dimensional and two-dimensional lattices of other magnetic or nonmagnetic materials [8-14]. Crystallization process is key factor which significantly impact on films’ quality. Authors fabricate double-layers films of different compositions and type of substrates. This work shows the investigation of deviations of films’ microstructure compare to its composition, substrates and growth conditions.
2. Experimental

Bi:IG Reactive ion beam sputtering method (RIBS) was used to fabricate Bi:IG double-layers at a URM 3-279.014 setup in oxygen–argon mixture using an ion-beam Kholodok-1 source. The films were deposited from ceramic target on gadolinium gallium garnet (GGG) substrates with crystallographic orientation of (111) or fused quartz SiO2 substrates [15–18]. Crystallization annealing at air (atmosphere pressure) and the temperature range from 650°C to 710°C and time period from 20 min to 60 min formed garnet phase. Different compositions of Bi:IG were used: G1 – Bi1.0Lu0.5Gd1.5Fe4.2Al0.8O12, G8 – Bi2.5Gd0.5Fe3.8Al1.2O12, GN – Bi2.8Y0.2Fe5O12.

All investigated samples have two layers (G1/G1, G1/G2 or G1/GN) with thicknesses of first G1-layer, second G1-, G8- and GN-layers of 80 nm, 230 nm, 195 nm and 177 nm, respectively. Two key process modes (methods) of crystallization annealing were carryout. Each layer of the structure were crystallized separately in both modes. Temperature of annealing depended on the composition and was 700°C, 680°C and 650°C for G1, G8 and GN, respectively.

First method (dipping), hereinafter referred "I", description:
- The sample was almost immediately dipped in a thermal field at the annealing temperature.
- Crystallization annealing of the sample occurred within 20 min after reaching thermodynamic equilibrium.

Second method (slow), hereinafter referred "II", description:
- The synthesis was carried out at very slow heating (heating rate of about 2-3°C / min) of the sample to the optimum temperature for a given composition and subsequent crystallization at a constant temperature for 60 min.

Crystallization features of double-layers films were revealed by surface structure which depends on process parameters (type of substrate, thickness of layers, temperature and time of annealing). Atomic force microscopy (AFM NTEGRA NT-MDT) was used to measure topography parameters of Bi:IG thin films. Active vibroisolation platform was used to protect the system from vibration noises. Average roughness of each film was measured to determine the process regime which allows to minimize this
parameter. Tapping mode of AFM was used for investigations because it allows one to get the best value of resolution and minimize the negative influence of the probe on the sample. In this mode, the cantilever console oscillates with self-resonant frequency and high amplitude (approximately 50-100 nm). At these amplitudes, the needle contacts the sample surface at the moment of maximum deflection of the console down from its equilibrium position, which significantly changes the frequency, phase and amplitude of its oscillations. A tapping mode has a higher horizontal resolution compared to the contact mode.

Cantilevers NSG10 (TipNano) were used with resonant frequency of 240 kHz and force constant of 11.8 Nm.

We used AFM images of 15x15 and 2x2 microns area for analysis. Parameters of height range P, root mean square roughness RMS and polycrystalline size AGS were determined. AGS was estimated by advanced watershed method.

Faraday rotation angle and Faraday hysteresis loops (FHLs) were measured by compensation method using automated magneto-polarimeter at the wavelength of 655 nm. Magneto-polarimeter includes the light source (semiconductor laser), polarizer, MO modulator, electromagnet, compensator, analyzer, photodetector, electronic system and a personal computer. Special holder of the sample and a Hall sensor to measure the intensity of magnetic field were placed in the gap of electromagnet. The range of available magnetic fields was from -5 to 5 kOe.

Faraday rotation angle $\alpha_F$, specific Faraday rotation $\Theta F$, coercivity $H_C$, squareness coefficient $K_S$, saturation magnetic field $H_S$ were determined from FHLs. To calculate the specific angle $\Theta F$, the total thickness of magnetic layers was taken into account. The film thickness h was calculated on the basis of sputtering velocity with control by optical (MII-4 microinterferometer) and AFM methods.

Table 1 lists the determined structural and magnetic parameters of samples.

| Composition & method | $\Theta_F$, $^\circ/\mu$m | RMS, nm | P, nm | $H_C$, Oe | $K_S$, rel. units |
|----------------------|---------------------------|---------|-------|-------------|-----------------|
| $Gd_3Ga_5O_{12}$     |                           |         |       |             |                 |
| G1/G1 I              | 0,97                      | 4,6     | 89    | 426         | 0,98            |
| G1/G1 II             | 1,01                      | 5,9     | 65    | 487         | 0,99            |
| G1/G8 I              | 2,22                      | 3,4     | 51    | 240         | 0,52            |
| G1/G8 II             | 2,22                      | 9,8     | 60    | 264         | 0,71            |
| G1/GN I              | 2,92                      | 7,8     | 52    | 230         | 0,24            |
| G1/GN II             | 5,25                      | 5,2     | 80    | 193         | 0,19            |
| $SiO_2$              |                           |         |       |             |                 |
| G1/G1 I              | 0,75                      | 4,6     | 47    | 384         | 0,84            |
| G1/G1 II             | 1,01                      | 12      | 142   | 501         | 0,91            |
| G1/G8 I              | 1,75                      | 9,6     | 119   | 389         | 0,66            |
| G1/G8 II             | 2,07                      | 5,9     | 65    | 469         | 0,68            |
| G1/GN I              | 2,92                      | 8,1     | 65    | 289         | 0,27            |
| G1/GN II             | 3,50                      | 9,1     | 117   | 241         | 0,19            |
3. Results

Figures 1 demonstrates the correlation of topography and FHLs of three types of samples. All samples, regardless of their composition, have polycrystalline surface after the crystallization. As it was noted earlier in our works, the processes of crystallization proceed differently on substrates which has the garnet structure and without [19,20]. Epitaxial growth of Bi:IG polycrystals occurs on garnet substrates or layers. But in case of SiO2 substrates garnet phase growth occurs by the mechanism of spontaneous crystallization. According to AFM data in both cases the film formation goes by an island mechanism. For the GGG substrate the growth of individual oriented polycrystals distributed evenly over the surface of the substrate is observed [21]. The typical size of polycrystals is the same as it shown in Figure 1. In the case of a SiO2 substrate, the formation of polycrystals begins spontaneously in some places on the surface of the substrate, and the film formation goes by combining micron sizes aggregates of grains. It is also seen by increasing the saturation fields of samples from GGG substrate to SiO2 (Figure 1., FHLs). Grains on SiO2 substrates have a larger anisotropy spread.

As a result, the film of G1 composition is formed on the surface of SiO2 only with a layer thickness bigger than 60-70 nm [22]. Garnet structures of the G8 and GN compositions are not formed on SiO2. So, the G1-layer with thickness of 80 nm was set for double-layer films. Its purpose is to be a seed for the crystallization of the second main layer G1, G8 or GN. These layers are considered by us to be functional of the two-layer film as a whole.

The structure of double-layer films depends on the initial structure of the sublayer G1. For instance, the surface of double-layer G1/G8 films demonstrates different features on GGG and SiO2 substrates, respectively. The surface of the films on GGG is characterized by a uniform distribution of crystallites. The grains are approximately the same size, even on a large-area frames. Average size of the crystallites is about 90-100 nm. The surface of the films on SiO2 shows the presence of regions with different crystal structures – regions with large crystallites of about 200-500 nm of a clear rectangular or triangular shape, smooth regions with small "round" crystallites of the same size or regions clearly being the boundaries of intergrown conglomerates. A fragment of a 2x2 µm smooth region is shown in Figure 1.

The most uneven structure is observed on SiO2 substrates samples at first annealing mode. The grains have the greatest size variation and even nano-cracks (Figure 1, c). Films are formed at the second annealing mode have more homogeneous structure on SiO2. The longer annealing time (second mode) leads to re-formation of centers of crystallization into a large smooth polycrystalline film. In contrast, an increase of annealing time leads to the formation of large crystallites and the heterogeneity of their distribution on the GGG. Figure 2 shows the surface structure of such films on GGG and the histograms of their grain size distribution. Topography comparison on GGG substrate compare to annealing method shown on Figure 3.

Depending on the second layer composition we vary the magnetic characteristics – the type of anisotropy. The films G1/G1 has the maximum squareness ratio $K_S = 0.99$ and low saturation field $H_S = 0.59-1.10$ kOe (Figure 1, FHLs). It confirms the implementation of «easy axis» anisotropy type. «Angle phase» and «easy plane» anisotropies are realized in G1/G8 ($K_S = 0.66, H_S = 0.75-1.34$ kOe) and G1/GN ($K_S = 0.27, H_S = 0.74-1.60$ kOe) films.

We should notice that studies other samples showed different results. The second annealing method led to the formation of a uniformly distributed polycrystalline structure for the SiO2 substrate and the formation of crystallites of various shapes for the GGG substrate.

So in order to synthesize a smoother film, we need to choose a faster synthesis mode (the first method) for growth on GGG substrates and a slower synthesis mode (the second method) for growth on SiO2 substrates. Table 2 shows the morphological parameters of all studied two-layer samples.
Figure 2. Grain analysis of garnet structure of different composition on GGG substrate. The films were annealed by second mode II.

Figure 3. Topography of garnet structure of different composition on GGG substrate. The comparison of films annealed by first I and second II mode.

However, these parameters showed a dependence on the crystallization method. Thus, the films G1/G1 and G1/G8 demonstrate more pronounced uniaxial anisotropy in the second annealing method. This may also be due to the diffusion of garnet-forming elements from the substrate during long-term annealing in the case of a GGG substrate (Table 1).

The decreasing values of the Faraday rotation for SiO2 films occurs quite strongly when the first mode is applied (Table 1).

For samples G1/G1 and G1/G8 trends in the magneto-optical and magnetic parameters coincide. The films on GGG at the first and second annealing modes are identical.

The difference in annealing modes is most clearly visible for samples on SiO2. For all types of such films, the second annealing mode is most advantageous.
For G1/GN the first annealing mode leads to a decreasing films’ quality for any substrate. This composition has the largest lattice parameter and its crystallization processes apparently require more specific conditions without a sharp change in the temperature gradient.

4. Conclusion

Thus, the influence of the crystallization annealing mode on the formation of the Bi:IG double-layers crystal structure is studied. Two methods, called briefly by the authors "dipping" and "slow", were compared. The both method is suitable for crystallization of Bi:IG films of composition G1/G1 and G1/G8 on GGG substrates. However, the second method is better suited for the formation of all investigated Bi:IG films on fused quartz SiO2 substrates which do not have the initial garnet structure and G1/GN regardless of the substrate type. It was found that smoother and more homogeneous samples on non-garnet substrates can be obtained using slow and prolonged annealing, while high-quality films on garnet substrates are formed with shorter heating and annealing.

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