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The Properties of Nanocomposite (BiDy)₃(FeGa)₅O₁₂:Bi₂O₃ Magneto-optic Garnet Films for Applications in Nanophotonics, Ultrafast Switching and Integrated Optoelectronics

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Abstract—We investigate the properties and applications of newly-developed RF-sputtered nano-composites with recordhigh magneto-optic quality (exceeding 25° at 532 nm and 42° at 635 nm). Bi-substituted dysprosium-gallium iron garnet layers with excess co-sputtered bismuth oxide content are demonstrated to possess very attractive optical and magnetic properties, which makes them suitable for novel magneto-optic and nanophotonic applications.

Index Terms—Magneto-optic materials, garnets, nonreciprocal effects, ultrafast switching, spatial light modulators, nanophotonics.

I. INTRODUCTION

The extra-ordinary optical and magneto-optical (MO) properties of Bi-substituted iron garnets continue to attract significant research interest worldwide from groups working in a wide range of application areas since the original discovery of giant Faraday effect in this class of materials [1]. This is due to the fact that Bi-substituted iron garnet (Bi:IG) materials outperform all other known transparent substances in terms of their MO figure of merit (defined as the doubled ratio between the specific Faraday rotation and the optical absorption coefficient). Bi:IG films possess moderately high absorption but, typically, a giant Faraday rotation in the visible range and are remarkably lossless in the near-infrared spectral region, yet still demonstrate world-record-high Faraday rotations (up to several thousand °/cm). For example, a specific Faraday rotation of 0.3 °/ μ m at 1300 nm wavelength in films of Bi_{1.56}Gd_{1.44}(FeAlGa)₅O₁₂ has been reported [2]. Bisubstituted iron garnets of different nominal compositions produced by a range of synthesis techniques have been studied and characterised extensively [2-9].

During the last several decades, Bi:IG thin films have been used in a number of application areas which belong to many different fields ranging from MO recording and imaging [7,11] to ultrafast switching [8], nanophotonics [9-10, 12-15], biomedical science [16], cold atoms manipulation, quantum information processing [17,18] and advanced optoelectronics [19,20].

The optical quality and magnetic properties of sputtered Bi:IG films (especially for large Bi substitutions) have so far been known to be rather difficult to control. In this paper, we report on the development of a new type of nanocomposite MO materials of composition $(BiDy)_3(FeGa)_5O_{12}:Bi_2O_3$ which demonstrate a record three-fold increase in MO quality factor defined as $(2\Phi/\alpha)$, where Φ is specific Faraday rotation and α is absorption coefficient, compared with usual RF-sputtered Bi:IG films [10]. The composite is represented by a matrix of bismuth oxide with embedded $(BiDy)_3(FeGa)_5O_{12}$ nanocrystals

of typical size 40 nm, as derived from x-ray diffraction (XRD) measurements.

II. THE CRYSTAL STRUCTURE AND MAGNETIC PROPERTIES OF NANOCOMPOSITE GARNET FILMS

Simultaneously with the large improvements in the MO figure of merit measured across the entire visible part of the garnets' transparency range, other significant advantages of nanocomposite garnet-Bi-oxide films have been found. The temperatures required to anneal the as-deposited sputtered films (crystallize them from the amorphous phase detected immediately after the film deposition) were found to be significantly lower for the best-performing nanocomposites than these required for processing typical sputtered garnet films of similar stoichiometry and properties. For example, running the nanocomposite crystallization processes at temperatures as low as 500-580 °C is clearly preferable to annealing typical Bi₂Dy₁Fe₄Ga₁O₁₂ layers at 680-700 °C, because the lower-temperature processes are compatible with a very wide variety of other material types (substrates, waveguides, other deposited layers) also present in any nanophotonic device or system under development.

Fig. 1 shows the x-ray diffraction data and pattern indexing results obtained from several types of newlygarnet-Bi-oxide nanocomposite developed MO materials. The excess off-stoichiometric bismuth oxide still remained in its amorphous phase after the hightemperature annealing processes and therefore did not show any x-ray diffraction peaks of its own. The bodycentered cubic structure of garnet nanocrystallites, which are embedded into the amorphous Bi-oxide matrix, has revealed itself unambiguously in the measured XRD patterns, allowing lattice constant determination. It was found, from the observed increases in the lattice constants of nanocomposite garnets compared with these of conventionallysputtered garnets of the same composition, that Bi substitutions were slightly higher in co-sputtered layers. The latter result was also confirmed by the observed increases in the specific Faraday rotation values of some co-sputtered nanocomposites. Two effects occurring simultaneously were found responsible for the record-high MO figures of merit measured in some co-sputtered garnet nanocomposites: the increase in the specific Faraday rotation caused by larger Bi substitutions, and the decrease in the visiblerange absorption coefficients across the spectrum, caused by the transparent property of excess Bi₂O₃ surrounding the garnet nanocrystallites.

Fig. 2 illustrates the attractive magnetic memory and switching properties observed in high-performance nanocomposite garnet-Bi-oxide materials compared to a typical magnetic hysteresis loop of sputtered $Bi_2Dy_1Fe_4Ga_1O_{12}$ layers.



Fig. 1. XRD datasets of several sputtered garnet and co-sputtered garnet-Bioxide layer types. All films were deposited onto Corning 1737 glass substrates. Specific Faraday rotations measured at 532 nm in all crystallized materials are shown together with the corresponding annealing regime data.



Fig. 2. Measured hysteresis loop of Faraday rotation (at 532 nm) of typical $Bi_2Dy_1Fe_4Ga_1O_{12}$ films post-deposition-annealed at 700 °C (a) and that of two different garnet-oxide nanocomposite layers annealed at temperatures between 560-580 °C (b). All films were deposited onto GGG (111) substrates.

Figure 3 shows the microstructure of a typical $Bi_2Dy_1Fe_4Ga_1O_{12}$ layer which was annealed at 700 °C after being sputtered in low-pressure (1 mTorr) pureargon atmosphere onto a GGG (111) substrate kept at 250 °C during the deposition process. Densely-packed small-size (about 40-50 nm) grains of garnet as well as the grain boundaries can be seen in this high-resolution transmission electron microscopy (TEM) image.



Fig. 3. A cross-section microstructure image (TEM) of a garnet layer on a GGG substrate (image is courtesy of Electron Microscope Unit, the University of New South Wales, Sydney, Australia).

The existence of thin (of about 15 nm thickness) interface layers between the substrates and the deposited garnet layers revealed during our microstructural investigations has important implications for the design of magnetic multilayer structures and magnetic photonic crystals, which often contain very thin (quarter-wavelength-thick) magnetic material layers. We intend to study the magnetic properties, microstructure details and the chemical composition of ultra-thin (sub-50nm) garnet and garnet-Bi-oxide layers in the future, in order to clarify their properties.

The XRD studies of our garnet-Bi-oxide nanocomposite layers have revealed essentially the same average garnet crystallite size (about 40 nm from using the Sherrer's formula and the diffraction peaks' linewidth data) as that typically observed in the cross-TEM images of the sectional crystallized Bi₂Dy₁Fe₄Ga₁O₁₂ layers which didn't contain extra cosputtered Bi₂O₃. This suggests that nanocomposite layers contain either the isolated nanocrystallites of garnet surrounded by the amorphous Bi₂O₃ matrix, or, more likely, the nanoclusters of multiple garnet nanocrystallites submerged within the matrix mainly composed of the amorphous Bi₂O₃. The latter preliminary conclusion will have to be confirmed directly using further high-resolution TEM studies. The comparison of magnetic hysteresis loops and the magnetic switching properties of the typical garnets and nanocomposite garnet-oxide layers shows substantial differences in the switching field values as well as in the magnetization process dynamics. In our opinion, the comparison of these properties supports the view that garnet nanocrystallites (or groups of these) within the nanocomposite garnet layers are separated geometrically.

III. APPLICATIONS IN MO IMAGING AND ULTRAFAST SPATIAL LIGHT MODULATORS

A. MO imaging using sputtered nanocomposite garnet-oxide films

High-contrast MO images of digital data recorded onto magnetic media such as floppy disks were memorized by our garnet nanocomposite films after a brief surface contact with these media and later revealed using polarization microscopy (Fig. 4). The excellent remanence (magnetic memory properties) of our films makes it possible to permanently imprint the images of bits recorded onto almost any perpendicularlymagnetized magnetic medium into the film's magnetic domains patterns.



Fig. 4. Applications of MO garnet nanocomposite films in MO imaging and digital data recovery from magnetic media. (a) A visible-light transmissionmode polarization microscope image of data tracks recorded on a 1.44 MB diskette obtained after a brief mechanical contact between the imaging film and disk surface; (b) a UV (365 nm) polarization microscopy image of a credit card's data-stripe section, imprint-memorised by a MO film with submicron-size domains that enable submicron-resolution MO imaging potential.

After storing the digital media information within the film's own domains magnetization distribution, highcontrast MO images can later be generated in either the transmission or reflection mode, without the necessity to keep the magnetized objects in contact with the imager films during inspection. Using UV polarization microscopy and MO films with very small domain size, ultra-high-resolution MO images can in principle be generated, which is expected to be useful for forensic data recovery from high-density magnetic recording media.

B. MO spatial light modulators

Owing to the attractive magnetic switching properties of sputtered MO composite films possessing very high specific Faraday rotation in the visible range, the development of current-driven pixelated MO devices for ultrafast image generation (transparencies and spatial light modulators) is possible. These devices will be composed of magnetically-isolated pixel arrays in which the (remnant) magnetization states of each individual pixel needs to be controlled individually. The pixel magnetization states can be controlled by a grid of conductors deposited, for example, by ion-assisted Pt deposition within the focused ion beam (FIB) milled grooves (Fig. 5) formed in garnet films.



Fig. 5. (a) a 5x5 matrix of magnetically-isolated MO pixels formed within a sputtered garnet layer using FIB milling; (b, c) polarization microscopy images (obtained several degrees away from the light extinction condition, each side away from extinction) of a 2x2 MO pixel array with magnetically-isolated pixels of size 20x20 μ m proposed for ultrafast MO spatial light modulators and image generation devices.

Experimental developments are being carried out, confirming that it is necessary to mill the grooves down to the substrate surface level in order to achieve complete magnetic separation of pixels formed in our films and develop practical arbitrarily-addressable magnetic pixel arrays which will find applications in ultrafast spatial light modulators and image recognition systems.

IV. CONCLUSION

We have discussed the magneto-optical properties of nanocomposite films of composition type $(BiDy)_3(FeGa)_5O_{12}:Bi_2O_3$ developed using RF magnetron sputtering. We have shown that such MO films can possess very high MO performance and very attractive magnetic properties, making them attractive components for applications in nanophotonics, ultrafast switching and integrated optoelectronics.

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