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## Recent developments in magneto-optic garnet-type thin-film materials synthesis

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### Abstract

Magneto-optic (MO) garnets are used in a range of applications in nanophotonics, integrated optics, communications and imaging. Bi-substituted iron garnets of different compositions are the most useful class of materials in applied magneto-optics due to their excellent MO properties (large Faraday effect) and record-high MO figure of merit among all semi-transparent dielectrics. It is highly desirable to synthesise garnets which possess simultaneously a high MO figure of merit and large uniaxial magnetic anisotropy. However, the simultaneous optimization of several material properties and parameters can be difficult in single-layer garnet thin films, and it is also challenging to prepare films with high bismuth content using physical vapor deposition technologies. To meet the current challenge of developing next-generation functional MO materials, we design, develop and demonstrate the functionality of new magnetostatically-altered all-garnet multilayer heterostructures using two different garnet materials of dissimilar anisotropy types (out-of-plane and almost-in-plane). The multilayer structures possess simultaneously a high MO figure of merit and large uniaxial magnetic anisotropy together with low coercivity, if each of the layers is optimized in composition and annealed correctly. We prepare thin-film heterostructures by sandwiching a MO garnet layer with almost in-plane magnetization in-between two MO garnet layers with out-of-plane magnetization using RF sputtering. We apply customised high-temperature oven annealing processes (optimized in temperature and process durations after running many trials) for the as-deposited (amorphous) garnet multilayers to obtain the crystalline garnet phase in every layer. These structures then possess simultaneously a high optical/MO quality and low coercivity, which is very attractive for the development of magnetic photonic crystals, sensing devices and ultra-fast switches. Based on Bi-substituted ferrite garnets grown on garnet substrates, this new and unique method for the development of new magnetic materials, enables customized magnetic properties to be attained, and can be used to develop novel types of synthetic garnet materials.

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**Keywords:** MO garnets, nanocomposites, co-sputtering, multilayer heterostructures, nanophotonics, integrated optics, magneto-photonic crystals, magneto-plasmonic crystals.

## 1. Introduction

Bi-substituted iron garnets of different composition types are the most useful class of functional materials in applied magneto-optics, due to their excellent MO properties (large Faraday rotation) and record-high MO figure of merit among all semi-transparent dielectrics. These materials can possess attractive magnetic properties and high specific Faraday rotation, if the deposited layers contain a high volumetric fraction of the garnet phase with good surface quality and microstructure. Numerous research studies have been conducted to understand the relationship between the MO properties (Faraday and Kerr rotation) and the bismuth content (substitution level, the number of Bi atoms per formula unit) in each garnet-type material system. A strong dependence of Faraday rotation (FR) on the level of bismuth substitution in thin film materials has been seen in garnet films developed using liquid-phase epitaxy (LPE). The extrapolated value of the specific FR of the fully bismuth-substituted  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  has been evaluated (based on these investigations) to be about  $6^\circ/\mu\text{m}$  at the wavelength of 633 nm [1]. Recently, electron cyclotron resonance (ECR) sputtered  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  films have been shown to demonstrate the specific Faraday rotation of about  $8.4^\circ/\mu\text{m}$  at 633 nm [2, 3]. LPE films having a record-high Bi-content demonstrated a linear growth of specific Faraday rotation with increasing bismuth content, which was obtained through compositional changes from  $\text{Lu}_3\text{Fe}_5\text{O}_{12}$  to  $\text{Bi}_{2.4}\text{Lu}_{0.6}\text{Fe}_5\text{O}_{12}$  [4]. In order to investigate the source of such disagreements between the measured properties of highly Bi-substituted garnets and theory predictions, and also to try and increase the number of bismuth atoms per unit formula in our sputtered films, we have synthesized a range of co-sputtered nano-composite garnet-type materials. In particular, we have prepared garnet-type composite thin films by co-sputtering from two separate ceramic (oxide-mix-based) targets using either two different garnet-stoichiometry targets or a garnet-type target co-deposited with an extra oxide [5,6]. The flexibility of the co-sputtering approach has enabled many final stoichiometry variations within nano-composites by simply varying the RF power levels applied to the sputtering targets. It has also been found that an important feature of RF sputtering deposition relevant to garnet materials synthesis is the ability to transfer relatively unchanged target stoichiometries to the films grown on various substrate types (garnet or glass) which allows growth of garnet-type films with bismuth substitution levels approaching three atoms per formula unit.

In this paper, we report on the successful synthesis of several new types of high-performance MO garnet-type nano-composite and heterostructure-type materials (obtained in the form of single-layer and multilayer thin films) of potential significance in applied magneto-optics which exhibit very promising properties suggesting suitability for a wide range of applications in integrated optics such as development of next-generation magnetic photonic crystals (MPC), magneto-plasmonic crystals, MO transparencies, ultra-fast switching and sensing devices.

## 2. Materials synthesis and related process parameters

The synthesis of thin-film garnet-type nano-composites and fabrication of all-garnet multilayer structures required optimization of several technological processes ranging from the sputtering target stoichiometry selection, substrate-cleaning techniques, RF sputtering deposition processes to the annealing crystallization processes (which needed to be fine-tuned for crystallizing each material type).

### 2.1. RF magnetron co-sputtering technology

All garnet and garnet-oxide composite thin films and all-garnet multilayer heterostructures were deposited using RF magnetron sputtering in pure argon plasma. RF magnetron sputtering is an enhanced method that allows thin film deposition at low operating pressures resulting in high quality thin films [4, 7-10]. In order to deposit thin-film garnet layers onto the substrates using sputtering technology, the following parameters needed

to be optimized: (i) target material stoichiometry and phase content, (ii) RF power densities applied to each target, (iii) substrate temperature, (iv) target-to-substrate distance, (v) deposition rate and argon pressure and (vi) the substrates stage rotation rate.

The composition adjustments in garnet-oxide composite co-sputtered derivatives were performed by using variations of RF power levels applied to the separate targets. Individually measured partial deposition rates from each target were used to calculate the volumetric concentration of excess oxides ( $\text{Bi}_2\text{O}_3$  or  $\text{Dy}_2\text{O}_3$ ) content or the excess dissimilar-garnet content introduced into the composite film volume as described in Ref. [5]. Fig.1 shows the schematic diagram of the co-sputtering system geometry and the related processing workflow diagram used to obtain garnet-type nano-composite films in polycrystalline phase.

The multilayer structures were prepared in a single-run deposition process using two sputtering targets of typical Bi-substituted iron garnets having dissimilar magnetic behavior types. Both sputtering targets used had a rather high bismuth substitution level to maximize specific Faraday rotation. The main process-related parameters used to prepare the garnet-oxide, garnet-garnet nanocomposites and also all-garnet heterostructures are summarized in Table 1. RF magnetron sputtering and co-sputtering processes are particularly attractive for multiple emerging applications of garnets for the following reasons: 1) good control over film layer stoichiometry and layer thickness is possible, and 2) sputtering is very compatible with multiple substrate types and other planar technology-related processes, thus enabling future on-chip integration of non-reciprocal components.

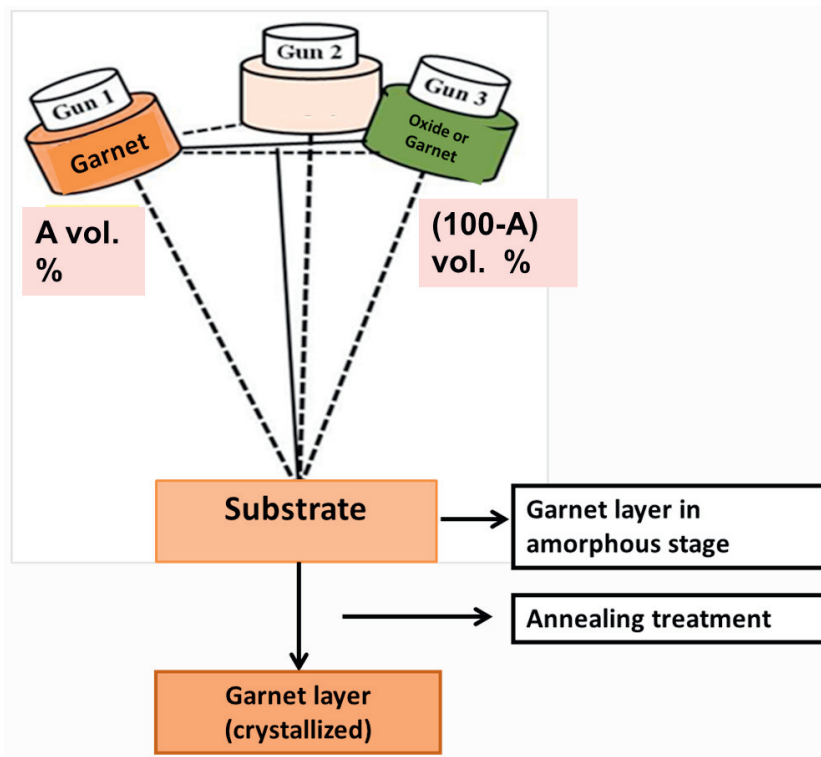


Fig.1. Schematic diagram of the co-sputtering system geometry and related processing workflow used for obtaining garnet-type co-sputtered nano-composite thin films.

Table 1. Summary of the sputtering process parameters used to deposit garnet-type composite thin films and the all-garnet multilayer structures.

Process parameters /Composition type	(BiDy) <sub>3</sub> (FeGa) <sub>5</sub> O <sub>12</sub> :Bi <sub>2</sub> O <sub>3</sub>	(BiLu) <sub>3</sub> (FeAl) <sub>5</sub> O <sub>12</sub> :Bi <sub>2</sub> O <sub>3</sub>	Bi <sub>2</sub> Dy <sub>1</sub> Fe <sub>4</sub> Ga <sub>1</sub> O <sub>12</sub> :Bi <sub>3</sub> Fe <sub>5</sub> O <sub>12</sub>	Bi <sub>3</sub> Fe <sub>5</sub> O <sub>12</sub> :Dy <sub>2</sub> O <sub>3</sub>	Bi <sub>2</sub> Dy <sub>1</sub> Fe <sub>4</sub> Ga <sub>1</sub> O <sub>12</sub> / Bi <sub>1.8</sub> Lu <sub>1.2</sub> Fe <sub>3.6</sub> Al <sub>1.4</sub> O <sub>12</sub> / Bi <sub>2</sub> Dy <sub>1</sub> Fe <sub>4</sub> Ga <sub>1</sub> O <sub>12</sub>
Sputtering target stoichiometry – oxide-mixed garnet targets	Bi <sub>2</sub> Dy <sub>1</sub> Fe <sub>4</sub> Ga <sub>1</sub> O <sub>12</sub> , Bi <sub>2</sub> Dy <sub>1</sub> Fe <sub>4.3</sub> Ga <sub>0.7</sub> O <sub>12</sub>	Bi <sub>1.8</sub> Lu <sub>1.2</sub> Fe <sub>3.6</sub> Al <sub>1.4</sub> O <sub>12</sub>	Bi <sub>3</sub> Fe <sub>5</sub> O <sub>12</sub>	Bi <sub>3</sub> Fe <sub>5</sub> O <sub>12</sub>	Bi <sub>2</sub> Dy <sub>1</sub> Fe <sub>4</sub> Ga <sub>1</sub> O <sub>12</sub> & Bi <sub>1.8</sub> Lu <sub>1.2</sub> Fe <sub>3.6</sub> Al <sub>1.4</sub> O <sub>12</sub>
Additional oxide or garnet-stoichiometry oxide-mix	Bi <sub>2</sub> O <sub>3</sub> (4-50 vol. %)	Bi <sub>2</sub> O <sub>3</sub> (4-20 vol. %)	Bi <sub>2</sub> Dy <sub>1</sub> Fe <sub>4</sub> Ga <sub>1</sub> O <sub>12</sub> (20-25 vol. %)	Dy <sub>2</sub> O <sub>3</sub> (2.7-20 vol. %)	NA
Base pressure (Torr)	5-6 × 10 <sup>-6</sup>	5-6 × 10 <sup>-6</sup>	5-6 × 10 <sup>-6</sup>	4-5 × 10 <sup>-6</sup>	5-6 × 10 <sup>-6</sup>
Argon (Ar) pressure	1-2 mTorr	1-2 mTorr	1-2 mTorr	2 -3 mTorr	1-2 mTorr
Substrate stage temperature (°C)	250	250	250	Room Temperature (21 °C)	250
Substrate stage rotation rate (rpm)	36-38	36-38	36-38	51-53	40-44
Annealing processes	620-700 (±5) °C for 1-3 h	560-630 (±5) °C for 1-10 h	510-550 (±10) °C for 1-3 h	550-630 (±5) °C for 1-3 h	600-630 (±10) °C for 1-6 h

## 2.2. Annealing process optimization

Neither the composite garnet-type films nor the multilayer structures showed any MO effects immediately after the deposition processes because of being in an amorphous phase after having been deposited at relatively cold substrate temperatures not exceeding 250 °C. To achieve good crystallinity in the garnet phase, the as-deposited thin film layers needed to be annealed, with each material system type requiring its own, fine-tuned annealing temperatures and durations. The optimized annealing regimes were also found to be dependent on the substrate type. By using a conventional high-temperature oven with air atmosphere to anneal all garnets and garnet-type thin film materials synthesized, we were able to find the optimized annealing regimes for each material type through multiple annealing trials followed by optical and MO characterization.

### 2.3. Thin film characterization

The annealed high-quality films (as judged by the best-obtained specific Faraday rotations at 532 nm and 635 nm) of nanocomposite-type and multilayer-type garnets were characterized using a Beckman Coulter DU 640B UV/Visible spectrophotometer, transmission-mode polarization microscope and a Thorlabs PAX polarimeter system used in conjunction with a custom-made calibrated electromagnet. The physical thicknesses of all films were first measured during the deposition process and reconfirmed using specialized custom-developed thickness-fitting software as well as the measured optical transmission spectra of both the as-deposited and post-annealed layers. Several crystallized composite films of type  $(\text{Bi}_3\text{Fe}_5\text{O}_{12}\text{-Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12})$  were also characterized using magnetic circular dichroism (MCD) spectroscopy. Hysteresis loops of Faraday rotation measured using an electromagnet revealed important details related to the magnetic switching behaviour, anisotropy type and magnetization vector direction in all materials synthesized.

## 3. Results and discussion

### 3.1. $(\text{BiDy})_3(\text{FeGa})_5\text{O}_{12}:\text{Bi}_2\text{O}_3$ composites

Highly Bi-substituted garnet-bismuth oxide composites (having between 4-50 vol.% of excess  $\text{Bi}_2\text{O}_3$  content) were prepared in low-pressure pure-argon plasma atmosphere inside the sputtering chamber using a substrate temperature of 250 °C. Many optimally-annealed garnet-type composite films demonstrated very high specific Faraday rotations (values are summarised in Table 2) simultaneously with low optical losses across most of the visible and near-IR spectral regions, leading to a record-high MO figure of merit at 635 nm. The MO performance of the co-sputtered garnet type films of this composition type was found to be significantly dependent on the volumetric concentration of excess  $\text{Bi}_2\text{O}_3$  added during the deposition processes. The best-obtained films of this type crystallized into body-centered cubic (bcc) lattice structure type and had a lattice parameter of 12.563 Å. The composition stoichiometry achieved in these films was close to  $\text{Bi}_{2.4}\text{Dy}_{0.6}\text{Fe}_4\text{Ga}_1\text{O}_{12}$ . Nearly square-shaped hysteresis loops of Faraday rotation (measured using 532 nm polarised light source) were observed in all garnet-oxide composite films and examples are shown in Fig. 2. The coercive force and switching field values were found to vary with the variations in extra  $\text{Bi}_2\text{O}_3$  content introduced into the composite films which confirmed the possibility of engineering the magnetic properties for applications in ultrafast switches and MO light modulators [6, 10]. The best achieved Faraday-effect magnetic field sensitivity (the ratio of increments in the Faraday rotation to the magnetic field strength increments) was about 38 °/(cm·Oe) at 635 nm (a summary of best achieved MO properties is given in Table 2). These excellent optical and MO property combinations make these composites very attractive for several applications in integrated optics and photonics such as nano-structured polarization controllers for communication-band wavelengths, MO visualizers for security and digital forensics and MO spatial light modulators for data processing.

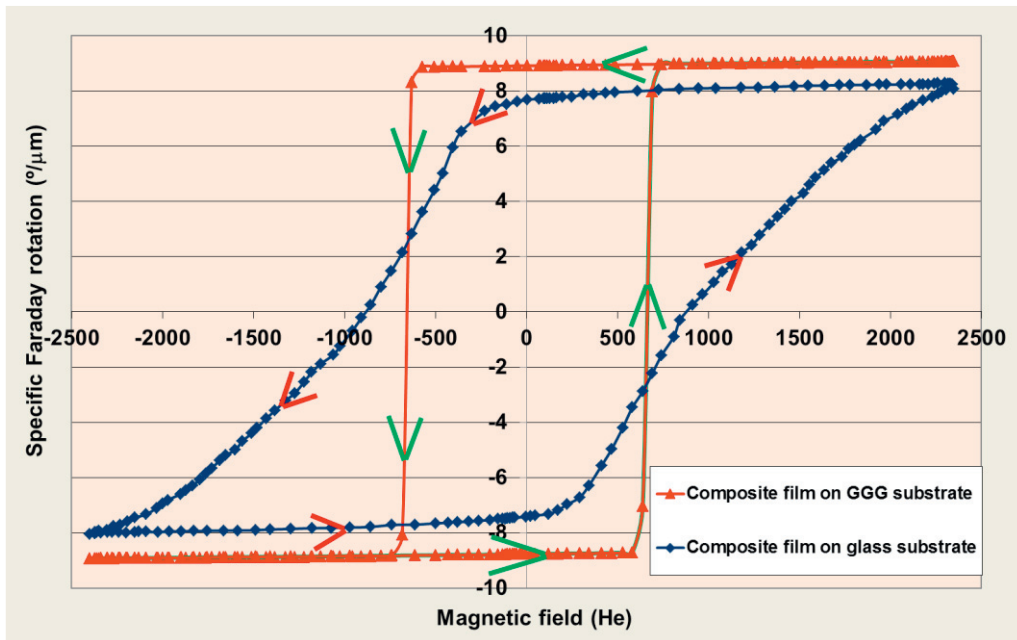


Fig.2. Hysteresis loops of specific Faraday rotation measured in  $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}:\text{Bi}_2\text{O}_3$  (about 22 vol. % of excess  $\text{Bi}_2\text{O}_3$ ) composite-type garnet film (1.1  $\mu\text{m}$  thick) grown on a GGG (111) substrate (red colour), and  $\text{Bi}_2\text{Dy}_1\text{Fe}_{4.3}\text{Ga}_{0.7}\text{O}_{12}:\text{Bi}_2\text{O}_3$  composite-type garnet film (having an estimated 24 vol. % of excess  $\text{Bi}_2\text{O}_3$ ) grown on a Corning 1737 glass substrate (blue colour). Films on glass substrates showed a different dynamics of the magnetization switching process in comparison with other films from the same batch sputtered onto GGG (111) substrates. The substrate type (glass or GGG) clearly influenced the magnetization switching processes, which is likely related to microstructural differences in layers, and this further can be illustrated by X-ray diffraction data obtained by us previously and presented in [10]

### 3.2. $(\text{BiLu})_3(\text{FeAl})_5\text{O}_{12}:\text{Bi}_2\text{O}_3$ composites

RF magnetron sputtered Bi-substituted aluminium-doped lutetium iron garnet thin film materials are very important for the applications that require high-quality thin-film magnetics possessing low coercive force but high specific Faraday rotations. Thin film garnet materials of this class possess an almost in-plane magnetization direction (as confirmed by the obtained linearity of response in their hysteresis loop of Faraday rotation). Fig. 3 shows the measured hysteresis loop of Faraday rotation in annealed composite thin films of composition type  $(\text{BiLu})_3(\text{FeAl})_5\text{O}_{12}:\text{Bi}_2\text{O}_3$  (12.5 vol. %) shown in comparison with that obtained from a typical  $(\text{BiLu})_3(\text{FeAl})_5\text{O}_{12}$  garnet film (without extra oxide dilution) deposited onto a glass substrate. The additional  $\text{Bi}_2\text{O}_3$  content reduced the specific Faraday rotation slightly but also reduced the coercive force and the magnetization saturation field values. The variations in the coercive force values and the optical absorption coefficients were found to be dependent on the excess volume fraction of  $\text{Bi}_2\text{O}_3$ . The best achieved quality factor (MO figure of merit  $2\Theta_F/\alpha$ , where  $\Theta_F$  is the specific Faraday rotation and  $\alpha$  is optical absorption coefficient) in this composition type exceeded 50 degrees (measured at 635 nm) [11].

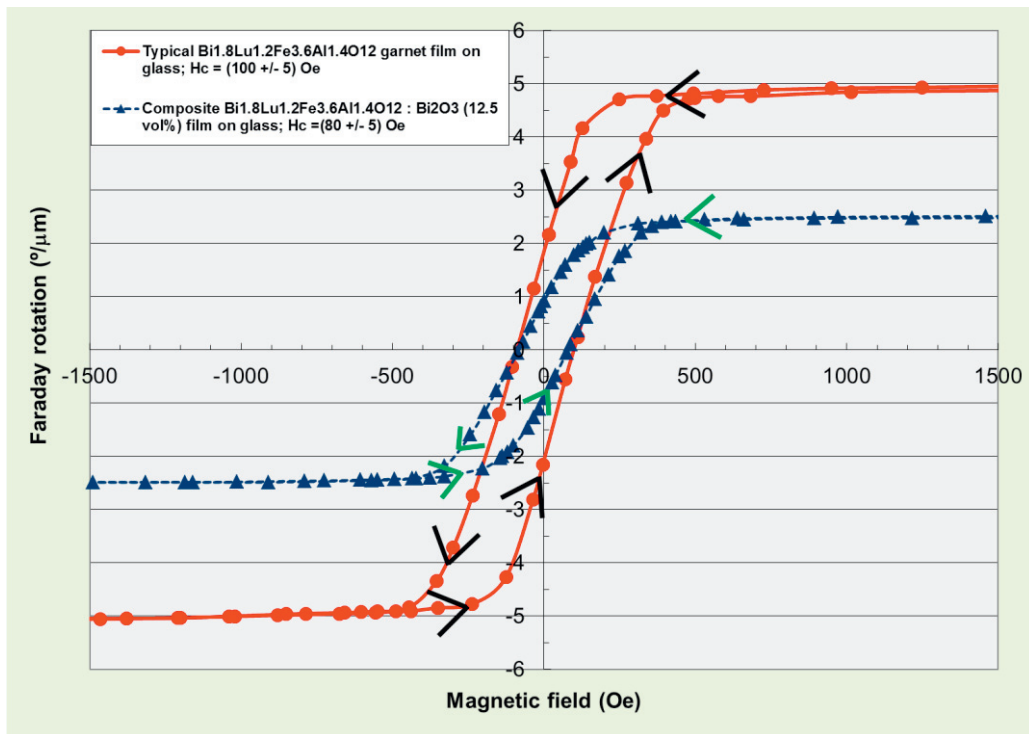


Fig.3. Hysteresis loops of Faraday rotation measured in a garnet-type composite film  $(\text{BiLu})_3(\text{FeAl})_5\text{O}_{12}:\text{Bi}_2\text{O}_3$  (12.5 vol. %) prepared on a glass (Corning Eagle XG) substrate (blue-coloured trace), compared with that of a typical garnet film of composition type  $\text{Bi}_{1.8}\text{Lu}_{1.2}\text{Fe}_{3.6}\text{Al}_{1.4}\text{O}_{12}$  deposited onto glass substrate (red-coloured trace).

### 3.3. All-garnet nano-composite thin film materials

The co-deposited all-garnet garnet-mix-type films were synthesized in order to approach the ultimate bismuth substitution level without using a single-target  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  sputtering process which we found to result in films not crystallizing into garnet phase. These films exhibited rather high specific Faraday rotation (around  $8.8 \text{ }^\circ/\mu\text{m}$  in a co-sputtered garnet mix composed of 75 vol.%  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  and 25 vol.% of  $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$  annealed at  $540 \text{ }^\circ\text{C}$ ) at  $532 \text{ nm}$  (compared to around  $6.9 \text{ }^\circ/\mu\text{m}$  measured in typical thick  $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$  films which usually required annealing at temperatures greater than  $650^\circ\text{C}$ ) simultaneously with good surface quality. These mixed-garnet films also showed strong remnant magnetization (memory behaviour observed during the specific Faraday rotation measurements) with magnetization direction being perpendicular to the film plane. Nearly-square shape of hysteresis loop of Faraday rotation was observed in this thin film type (shown in Fig. 4) with a coercive force value of about  $250 \text{ Oe}$  and the saturation field of near  $700 \text{ Oe}$ . The MCD (magnetic circular dichroism) measurements confirmed the successful synthesis of high-performance garnet-type MO material, which also exhibits a very significant “red-shift” in their main MO transitions that can be a very useful phenomenon for engineering garnets with very large Faraday rotation [12], making this garnet-type material attractive for the development of next-generation magnetic photonic crystal structures.



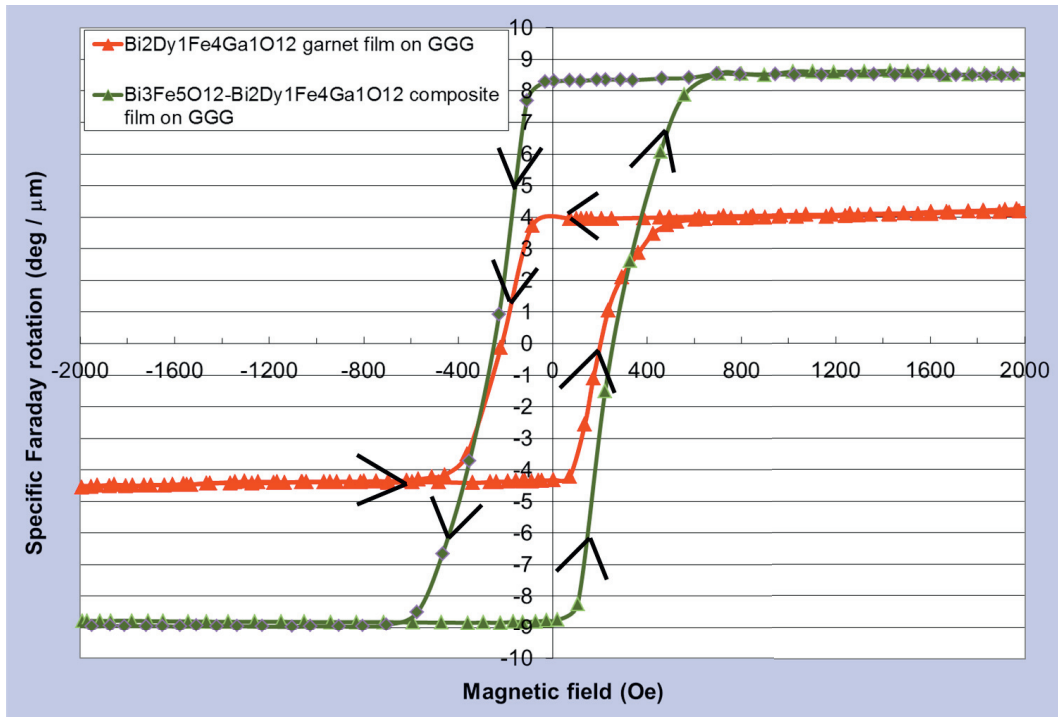


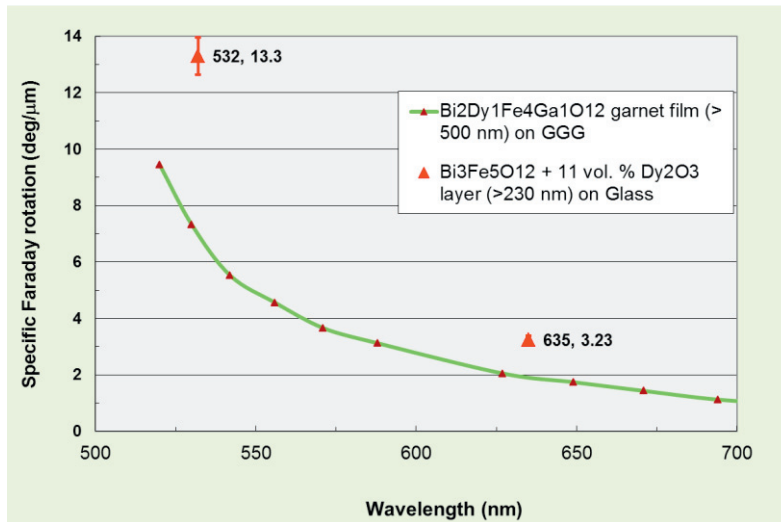
Fig.4. Hysteresis loop of Faraday rotation measured in an annealed garnet-type co-sputtered nano-composite ( $\text{Bi}_3\text{Fe}_5\text{O}_{12}$ - $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$ ) film of 1  $\mu\text{m}$  thickness prepared on a glass substrate (green-coloured trace) presented in comparison with that obtained in a typical  $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$  garnet film of the same thickness (red-coloured trace).

Magnetic circular dichroism (MCD) characterisation results of highly bismuth-substituted garnet-garnet composites of type  $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$  -  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  showed that a significant “red shift” of MO characteristics occurred in this material type (as was reported by us in [12]), which we believe is related to the answer to the question raised by us in the Introduction section – why strong increases in Faraday rotation (beyond the predicted 2deg/micron at 633 nm per formula unit of Bi) are observed near full bismuth substitution of 3 Bi atoms per formula unit

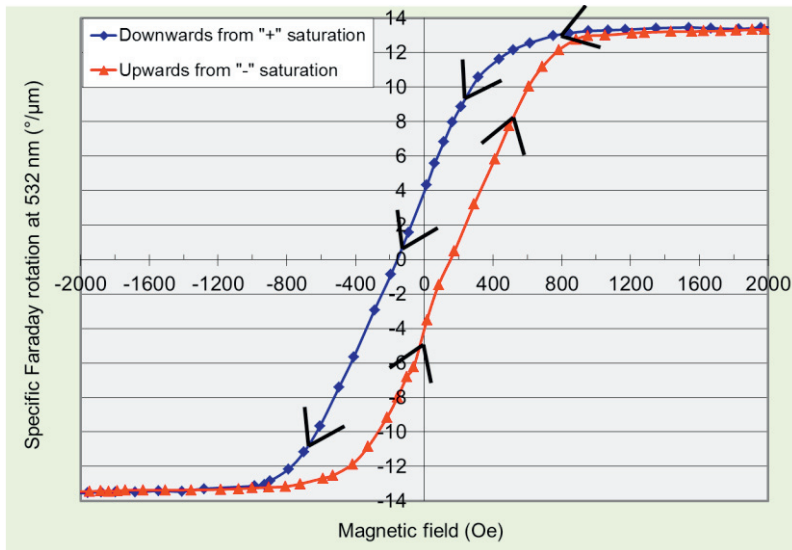
### 3.4. $\text{Bi}_3\text{Fe}_5\text{O}_{12}:\text{Dy}_2\text{O}_3$ composites

$\text{Bi}_3\text{Fe}_5\text{O}_{12}:\text{Dy}_2\text{O}_3$  (2.7-20 vol. %) garnet-type composite thin films (230-320 nm in thickness) were prepared using RF magnetron co-sputtering in an attempt to synthesize a garnet material with bismuth substitution level approaching 3 formula units. We were not able to synthesise  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  by means of post-deposition annealing of films sputtered from a target of this nominal stoichiometry, possibly due to other materials like  $\text{BiFeO}_3$  forming from amorphous-phase oxide mixes prior to the formation of garnet phase. The annealed garnet-dysprosium oxide composites showed very high Faraday rotation per film thickness (reaching 13.3  $^\circ/\mu\text{m}$  at 532 nm in a composition with 11 vol. % of excess dysprosium oxide) which also suggested the successful crystallization of a garnet composition having a large bismuth substitution level. Fig.5 (a) shows the specific Faraday rotation data points measured on a quarter-micron thick composite film ( $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  + 11 vol. %  $\text{Dy}_2\text{O}_3$ ) prepared on a glass substrate, compared with the specific Faraday rotation spectra obtained in a half-micron-thick typical  $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$  garnet film, while Fig. 5 (b) shows the obtained hysteresis loop of Faraday rotation measured in the same dysprosium oxide-diluted film. From the hysteresis loop shape, it is quite clear

that this material type possesses an almost in-plane magnetization direction. Note that on-going research is undertaken to further characterize this new type of garnet-oxide nanocomposites, and their microstructure and other properties will be reported elsewhere.



5(a)



5(b)

Fig.5. (a) Specific Faraday rotation data points (measured using 532 nm and 635 nm light) obtained from an annealed nanocomposite film of composition type  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$ : 11 vol. %  $\text{Dy}_2\text{O}_3$  prepared on a glass substrate. The films were subjected to annealing at  $530^\circ\text{C}$  for 1 h inside a conventional high-temperature oven in air atmosphere; (b) measured hysteresis loop of Faraday rotation of the same sample revealed that this nanocomposite film had a rather small coercive force value and had an almost in-plane magnetization.

### 3.5. Exchange-coupled all-garnet multilayer thin film heterostructures

Exchange-coupled all-garnet multilayer structures were fabricated using two record-performance highly-Bi-substituted iron garnet materials having very dissimilar magnetic behaviours (different magnetic anisotropy types, switching fields and saturation magnetizations) to investigate the special magnetic behaviour which results from heterostructure formation and magnetostatic coupling. The stoichiometries of the two bismuth-substituted iron garnet (Bi:IG) thin film materials used were  $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$ , and  $\text{Bi}_{1.8}\text{Lu}_{1.2}\text{Fe}_{3.6}\text{Al}_{1.4}\text{O}_{12}$ , which had previously been studied by our group in detail [6, 11, 13]. We had also reported previously that these materials can possess exceptional optical and MO qualities, together with good magnetic properties when mixed with extra co-sputtered bismuth oxide. All-garnet multilayer film structures were deposited onto paramagnetic substrates (GGG) by sandwiching a magneto-soft garnet material ( $\text{Bi}_{1.8}\text{Lu}_{1.2}\text{Fe}_{3.6}\text{Al}_{1.4}\text{O}_{12}$ ) in-between two magneto-hard materials ( $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$ ) of the same thicknesses and identified their optimized annealing process parameters (630-710 C and 1-10 hrs process duration). The annealed samples showed Faraday rotation exceeding  $\pm 3^\circ$  (total angle) at 532 nm, and the measured coercive force was around 100 Oe. The measured magnetic hysteresis loop is shown in Fig.6 [14]. The results obtained indicate that forming magnetostatically-coupled all-garnet multilayer heterostructures allows the engineering of new material systems that can inherit the best properties from their constituent layers having substantially different magnetic anisotropy types. The exchange-coupled all-garnet multilayer structures possessed high uniaxial magnetic anisotropy simultaneously with rather low coercivity, which is extremely useful for some specific applications in magnetically switchable devices for modulating light through Faraday effect.

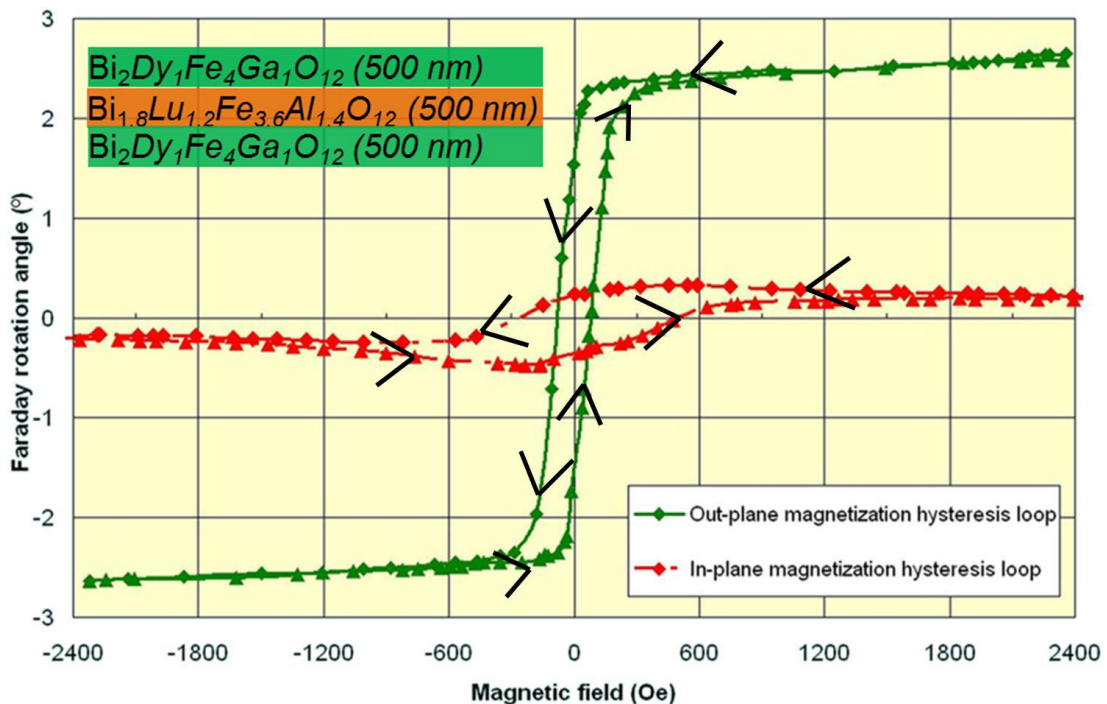


Fig.6. Magnetic hysteresis loops obtained from an all-garnet multilayer structure prepared on a GGG (111) substrate (annealed for 3h at 630 °C after the deposition) with an external magnetic field applied both in perpendicular direction (out-of-plane, green colour curve) and parallel (in-plane, red colour curve) with respect to the film plane of the multilayer structure. The thicknesses of each garnet layer within the entire heterostructure are also shown.

Table 2. Summary of the MO performance parameters achieved in each type of garnet-oxide or all-garnet composite thin films

Material composition	Best achieved Specific Faraday rotation ( $^{\circ}/\mu\text{m}$ )		Best achieved MO figure of merit ( $2*\Theta_F/\alpha$ , degrees)		Hysteresis loop properties	Best achieved Faraday-effect magnetic-field sensitivity at 635 nm ( $^{\circ}/\text{cm.Oe}$ )
	532 nm	635 nm	532 nm	635 nm		
$\text{Bi}_{2.0}\text{Dy}_{1.4}\text{Fe}_{3.6}\text{Ga}_{1.4}\text{O}_{12}:\text{Bi}_2\text{O}_3$ composites	9.8	2.6	29.5	42.5	Square-shaped loops, perpendicular magnetization & high remnant magnetization	38
$\text{Bi}_{2.0}\text{Dy}_{1.4}\text{Fe}_{3.6}\text{Ga}_{1.4}\text{O}_{12}:\text{Bi}_2\text{O}_3$ composites	10.12	1.66	29.2	27.8	Nearly-square shaped loops, perpendicular magnetization, high remnant magnetization	20
$\text{Bi}_{1.8}\text{Lu}_{1.2}\text{Fe}_{3.6}\text{Al}_{1.4}\text{O}_{12}:\text{Bi}_2\text{O}_3$ composites	6.25	1.19	22.17	50.22	Almost in-plane magnetized; very low coercive force values observed	148
$\text{Bi}_{2.0}\text{Dy}_{1.4}\text{Fe}_{3.6}\text{Ga}_{1.4}\text{O}_{12}:\text{Bi}_3\text{Fe}_5\text{O}_{12}$ composites formed by diluting $\text{Bi}_3\text{Fe}_5\text{O}_{12}$	8.25	1.52	NC	NC	Nearly square-shaped loops, perpendicular magnetization, high remnant magnetization	60
$\text{Bi}_3\text{Fe}_5\text{O}_{12}:\text{Dy}_2\text{O}_3$ composites	13.3	3.23	Min. 8.5	NC	Almost in-plane magnetization direction and low coercive force, good remnant properties ( $M_r \geq 0.7M_s$ )	213
$\text{Bi}_{2.0}\text{Dy}_{1.4}\text{Fe}_{3.6}\text{Ga}_{1.4}\text{O}_{12}/\text{Bi}_{1.8}\text{Lu}_{1.2}\text{Fe}_{3.6}\text{Al}_{1.4}\text{O}_{12}/\text{Bi}_{2.0}\text{Dy}_{1.4}\text{Fe}_{3.6}\text{Ga}_{1.4}\text{O}_{12}$ multilayer thin film heterostructures	Min. 2	0.7	NC	NC	Nearly-perpendicular out-of-plane magnetization and very low coercive force values	NC

\*NC-Not characterized / calculated

Recently, our network of research collaboration (distributed across Russia, Australia, India and Germany) has enabled the discovery of a very attractive new magneto-optic effect (longitudinal magneto-plasmonic intensity effect (LMPIE) that provides plasmon-mediated magneto-optic transparency modulation) using our RF-sputtered garnet materials of composition type  $\text{Bi}_2\text{Dy}_1\text{Fe}_4\text{Ga}_1\text{O}_{12}$  [15]. Additionally, a significant enhancement of transverse magneto-optic Kerr effect (TMOKE) has been demonstrated in magneto-plasmonic structures based on sputtered films of  $\text{Bi}_{1.8}\text{Lu}_{1.2}\text{Fe}_{3.6}\text{Al}_{1.4}\text{O}_{12}$ . The magnitude of TMOKE observed in magneto-plasmonic crystals reached about 13% [16]. We believe that our recently-synthesized garnet-type nanocomposites will be suitable for demonstrating further results in relation to achieving magnetic-field control over the propagation of light waves in magneto-plasmonic crystals.

#### 4. Conclusion

A number of garnet-type thin-film material systems have been synthesized using physical vapour deposition (RF magnetron sputtering) and characterised optically, magnetically and magneto-optically. Among the multiple bismuth-substituted rare-earth iron garnet compositions and garnet-oxide nanocomposites studied, several material systems have been found to possess unique combinations of physical properties of interest for use in non-reciprocal integrated optics and magnetic switching applications. In particular, triple-layer all-garnet heterostructures composed of magnetic materials of dissimilar anisotropy types have displayed high MO quality, near-perpendicular magnetization and rather low coercivity. These extra-ordinary magnetic material systems are highly attractive for use in various existing as well as new and forward-looking applications in photonics, optoelectronics, magnetic photonic crystals and magnetoplasmonics.

#### 5. References

- [1] Zvezdin, A. K., and Kotov, V. A., 1997. *Modern Magneto-optics and Magneto-optical Materials*, Institute of Physics Publishing. Bristol and Philadelphia. (IOP Publishing Ltd.)
- [2] Hansen, P., Krumme, J. P., 1984. Magnetic and magneto-optical properties of garnet films, *Thin Solid Films* 114, pp. 69 – 107.
- [3] Adachi, N., Isarai, K., Ota, T., Ishiyama, K., Ota, H., Arai, K., 2010. Synthesis of completely substituted  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  garnet films by metal organic deposition technique, [http://www.crl.nitech.ac.jp/ar/2010/0104\\_crl\\_ar2010\\_paper.pdf](http://www.crl.nitech.ac.jp/ar/2010/0104_crl_ar2010_paper.pdf)
- [4] Krumme, J. P., Doormann V., Willich, J., 1985. Bismuth iron garnet films prepared by rf magnetron sputtering, *Journal of Applied Physics* 57, pp. 3885-3887.
- [5] Alam, Mohammad, 2012. High performance magneto-optic garnet materials for integrated optics and photonics. *Theses: Doctorates and Masters*. Paper 528. <http://ro.ecu.edu.au/theses/528>.
- [6] Vasiliev, M., Nur-E-Alam, M., Kotov, V. A., Alameh, K., Belotelov, V. I., Burkov, V. I., Zvezdin, A. K., 2009. RF magnetron sputtered  $(\text{BiDy})_3(\text{FeGa})_5\text{O}_{12}:\text{Bi}_2\text{O}_3$  composite garnet-oxide materials possessing record magneto-optic quality in the visible spectral region, *Optics Express*. 17, pp. 19520-19535.
- [7] Boudiar, T., Payet-Gervy, B., Blanc-Mignon, M. F., Rousseau, J. J., Berre, M. L., Joisten, H., 2004. Magneto-optical properties of yttrium iron garnet (YIG) thin films elaborated by radio frequency sputtering, *Journal of Magnetism and Magnetic Materials*, 284, pp. 77–85.
- [8] Johansson, P., Khartsev, S. I., Grishin, A. M., 2006. Comparison of  $\text{Bi}_3\text{Fe}_5\text{O}_{12}$  film giant Faraday rotators grown on (111) and (001)  $\text{Gd}_3\text{Ga}_5\text{O}_{12}$  single crystals, *Thin Solid Films*. 515, pp. 477 – 480.
- [9] Challeton, D., Bechevet, B., Rolland B., Armand, M. F., 1990. RF sputtering of Bi-substituted garnet films for magneto-optic memory, *Journal of Magnetism and Magnetic Materials*. 83, pp. 37-38.
- [10] Vasiliev, M., Nur-E-Alam, M., Perumal, P., Kotov, V., Alameh, K., Lee, Y. T., Lee, Y. P., 2011. Annealing behavior and crystal structure of RF-sputtered Bi-substituted dysprosium iron garnet films having excess co-sputtered bismuth oxide content, *Journal of Physics D: Applied Physics*, 44, p. 075002.
- [11] Nur-E-Alam, M., Vasiliev, M., Kotov, V., Alameh, K., 2011. Highly bismuth-substituted, record-performance magneto-optic garnet materials with low coercivity for applications in integrated optics, photonic crystals, imaging and sensing, *Optical Materials Express* 1, pp. 413-427.
- [12] Nur-E-Alam, M., Vasiliev, M., Alameh, K., 2012. “New Class of Garnet Nanocomposites for Use in Magnetic Photonic Crystals Prepared by RF Magnetron Co-sputtering,” *Proceedings of High-capacity Optical Networks and Enabling Technologies*, pp. 184-187.
- [13] Nur-E-Alam, M., Vasiliev, M., Alameh, K., Kotov, V. A., 2010. “High-quality RF-sputtered magneto-optic garnet films of  $\text{Bi}_{1.8}\text{Lu}_{1.2}\text{Fe}_{3.6}\text{Al}_{1.4}\text{O}_{12}$  with low coercivity for applications in integrated optics, imaging and sensing devices,” *Proceedings of High-capacity Optical Networks and Enabling Technologies*, pp. 62-66.

- [14] Kotov, V. A., Popkov, A. F., Soloviev, S. V., Vasiliev, M., Alameh, K., Nur-E-Alam, M., Balabanov, D. E., 2012. Magnetic heterostructures with low coercivity for high-performance magneto-optic devices, *Journal of Physics D: Applied Physics*, 46, p. 035001.
- [15] Belotelov, V.I., Kreilkamp, L.E., Akimov, I.A., Kalish, A.N., Bykov, D.A., Kasture, S., Yallapragada, V.J., Gopal, A. V., Grishin, A.M., Khartsev, S.I., Nur-E-Alam, M., Vasiliev, M., Doskolovich, L.L., Yakovlev, D.R., Alameh, K., Zvezdin, A.K., Bayer, M., 2013. Plasmon-mediated magneto-optical transparency, *Nature Communications*, 4, p. 2128.
- [16] Pohl, M., Kreilkamp, L.E., Belotelov, V.I., Akimov, I.A., Kalish, A.N., Khokhlov, N.E., Yallapragada, V.J., Gopal, A.V., Nur-E-Alam, M., Vasiliev, M., Yakovlev, D. R., Alameh, K., Zvezdin A. K., Bayer, M., 2013. Tuning of the transverse magneto-optical Kerr effect in magneto-plasmonic crystals, *New Journal of Physics* 15, 075024.