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ADVANCED MAGNETO-OPTICAL MATERIALS AND DEVICES

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by

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ABSTRACT

The magneto-optical materials with both high Faraday rotation and high transmittance capabilities are greatly desired in high speed switches, isolators, and visible imaging systems. In this thesis work, new magneto-optical materials that possess both high Faraday effect and high transmittance in the visible range of the spectrum were studied and synthesized. The work done includes material synthesis, material properties testing, optical and magneto-optical properties testing, switching speed testing, and theoretical investigations. Specifically, garnet films, iron borate crystals and nanoparticles, and garnet particle films were successfully synthesized and switches made of these new materials were designed and tested at the wavelength of 532 nm, which is at the shorter wavelength of visible spectrum and is also among to the most sensitive range of the normal human eye at night.

New Bismuth iron gallium garnet thin-films (Bi$_3$Fe$_4$Ga$_4$O$_{12}$, BIGG) have been successfully deposited on gadolinium gallium garnet substrates with a pulsed laser deposition technique in our lab. X-ray diffraction analyses have proven that the BIGG films are of good epitaxial quality with a lattice constant close to 12.61±0.01 Å. The bandwidth of BIGG’s transmittance spectrum has been extended and its left edge has been shifted about 50nm towards the shorter wavelengths relative to those of Bi$_3$Fe$_5$O$_{12}$ (BIG) films. The BIGG film is more transparent than a BIG film although BIGG’s Faraday rotation angle is slightly less than that of a BIG film. The figure of merit of the BIGG garnet film has reached 16.5°, which is about 1.8 times that of a typical BIG film.
Currently, the switches using BIGG films were tested and a 2.4 ns response time had been reached with a Φ1 mm circular aperture at the wavelength of 532 nm.

Iron Borate (FeBO₃) is another material that is far superior in terms of the transmittance in the visible spectrum at room temperature to most garnet materials. In this study, a flux sintering technique was used to grow highly perfect iron borate platelet-like single crystals. Our test showed that the green platelets we obtained have a (001) crystallization orientation. Our switching testing experiments showed that the speed of the switch using FeBO₃ platelets has reached 1.2ns with a Φ1 mm circular aperture at the wavelength of 532nm. The switching speed is high, but the Faraday rotation could not reach more than 5° because of the natural birefringence of the material. The FeBO₃ is one of the orthoferrites with a large natural birefringence for the light propagated along the magnetization direction. The effect of birefringence on Faraday rotation reduced the maximum obtainable rotation. In order to eliminate the birefringence and further improve the transmittance, a high energy ball-milling technique was used to synthesize FeBO₃ nanoparticles. Our numerical simulation shows the nanoparticles could eliminate the birefringence, and concurrently keep the intrinsic Faraday rotation. After milling and centrifuging, the FeBO₃ nanoparticles’ sizes reached down to 30nm so far. The nanoparticles are to be hot-pressed and re-formed into an orientation-randomized new crystal. It is expected that the nanoparticles approach will overcome the birefringence, improve the transmittance, and concurrently keep the same switching speed. Further tests need to be conducted on these FeBO₃ nanoparticles.

New Bi₁.₈Y₁.₂Fe₄Ga₁O₁₂ (BYIGG) nanoparticles were fabricated with coprecipitation and annealing processes. The advantages of these techniques are high
yield and relatively low cost. When substituting Ga by Fe, BYIGG nanoparticles improved the transmittance compared to Bi$_{1.8}$Y$_{1.2}$Fe$_5$O$_{12}$ (BYIG) nanoparticles. The BYIGG nanoparticles are more yellowish, while the BYIG nanoparticles are more reddish. With high energy ball-milling and spin-coating techniques, new BYIGG films were coated on a Corning 1737 glass substrates. Our test showed that the Faraday rotation angle was about 2.5° for a 500μm thick BYIGG film, and the transmittance at the wavelength of 532nm was about 1%. The figure of merit ($=\theta/\alpha$, where $\theta$ is Faraday rotation angle and $\alpha$ absorption coefficient) was found to be 2.2°, which is among the reported ones from 0.2° to 9.6°. It was observed that contamination from ball-milling process decreased the transmittance. Further experiments need to be conducted, and one more challenge is to eliminate the contamination.

The switches made of the above materials are tested for speed response. As we know, if the circuit response time is larger than the films/crystals’ response times, the speed tested is limited by the driving circuit board. For this reason, a RF circuit and a miniature coil were specially and successfully designed to generate a magnetic field about 400 Oe and a circuit rise time less than 1 ns.

In the theoretical study part of this dissertation, we proposed an new technique and successfully simulated an optimized aperiodic multilayer structure to enhance both the Faraday effect and the transmittance. Bi-substituted yttrium–iron–garnet films and dielectric SiO$_2$ films were used in our simulation at the wavelength of 532nm. One-dimensional magneto-optical aperiodic multilayer structures were theoretically studied and optimized by means of a genetic algorithm. For one structure we optimized,
Faraday rotation per magnetic material thickness ($\theta/d_M$) has been enhanced 13.6 times compared to a single magnetic layer, from $2.06^\circ/\mu\text{m}$ to $27.98^\circ/\mu\text{m}$. For another optimized structure, the total Faraday rotation angle and figure of merit are 3.7 and 2.5 times larger than those of a single magnetic layer, respectively. All these results prove the effectiveness of this newly developed optimization technique.
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1.1 Optical Switches

Optical switches are in great demand for a wide variety of applications, most prominently in optical communications. Current available optical switch mechanisms include micro-electromechanical systems (MEMS), acousto-optic methods (AO), liquid crystals (LC), thermal (optical) methods (TO), electro-optic effects (EO), and magneto-optic effects (MO).

MEMS are small integrated systems where micromachined mirrors are rotated and an optical signal is deflected whenever an electrostatic force is applied. Their fabrication processes involve modified semiconductor fabrication technology, molding and plating, wet and dry etching, electro discharge machining, and other technologies capable of manufacturing very small devices. MEMS switches are mostly used in the integrated circuits, and their fabrication processes are relatively complicated and costly.

Acousto-optic switches deflect light when the acoustic waves generate a diffraction grating in the AO crystal \[^1\]. Due to the limitation of the velocity of the acoustic wave of the crystal, AO switches are typically used as Q-switching components, modulators, and frequency control components in spectroscopy. Liquid crystal switches find wide applications in display devices, where Liquid crystals function by rotating the polarization direction of an incident light in the presence or absence of an electric field.
Thermal optical switches are typically based on Mach-Zehnder (MZ) schemes. Heaters are used to insert or extract boilable liquid into or out of channels that change the refractive index along the two optical paths. Similar to TO switches, MZ are typically used in Electro-optic switches. The refractive index or absorption coefficient of EO switches is changed by an applied electric field \(^2\) due to the Pockels effect, Kerr effect or Stark effect. Both TO and EO switches function by changing the phase difference between the two beams confined in waveguide; therefore, it is difficult for them to handle free space communications.

Magneto-optical switches, based on the Faraday effect or Kerr effect, change the polarization direction of the incident beam by an applied magnetic field. The nonreciprocal character of the magnetooptic Faraday effect is of great technical importance and is a necessary condition to construct devices such as optical isolators \(^3, 4\). In Faraday experiment, the rotation of the plane of polarization is proportional to the intensity of the component of the magnetic field in the direction of the beam of light and the coil generating the field could be made large. Therefore, Faraday switches can handle large beam cross sections. Magneto-optical switches using the Faraday effect are ideal for free space communications.

### 1.2 Magneto-optical Materials

Among magneto-optical materials, iron garnet crystals and films have a potential of tunable operating frequencies, high switching speed, good long-term reliability, and
high thermal stability[3]. However, most garnets with high Faraday rotation have a high absorption in the shorter wavelength range too. The Magneto-optical devices made of these garnet materials are commonly operated in the near infrared and infrared range. But garnets with low absorption and high Faraday effect in the shorter range of the visible spectrum are desirable in a variety of applications, such as night-vision goggles, undersea free space communication, and Q-switches.

In the late 1960’s, Bismuth-substituted garnets were developed and it was discovered that the substitution of bismuth in iron garnets produced a large increase in the Faraday rotation. Bismuth doped garnet crystals produced by a liquid phase epitaxial (LPE) technique are commonly used as isolators and sensors in optical communication systems[4-10]. To reach the highest Faraday rotation with doping of bismuth, pure Bismuth Iron Garnet (Bi$_3$Fe$_5$O$_{12}$ BIG) was in need. However, because it is not thermodynamically stable, BIG cannot be synthesized with the LPE technique. With either reactive ion beam sputtering (RIBS) or pulsed laser deposition (PLD) techniques, BIG thin films were successfully synthesized on suitable garnet substrates. The pioneer works on these BIG films were performed by Okuda in 1987[11, 12]. The PLD technique was developed later and used to synthesize garnet films because it offers exact transfer of stoichiometry with a surface quality of films. It was found that pure BIG reached giant Faraday rotation angle of 6-8°/µm at 633nm[13-15]. The giant Faraday rotation of BIG makes it a good candidate material in high speed free space communication systems.

To achieve angles of Faraday rotations as high as possible and film thickness as thin as possible in samples has been a major goal in the development of transparent magneto-optical materials[5, 16, 17]. In this thesis work, we focus on theoretical and
experimental investigations of different magneto-optical materials in the visible range of the spectrum. Specifically, several new magneto-optical materials were synthesized. These materials possess both high Faraday rotation and high transmittance in the visible range of the spectrum, and switches with these materials are designed with a high switching speed. To focus on the shorter wavelength of the visible range, a Nd:YAG laser with the wavelength of 532 nm was used in Faraday switch testing.

A great number of magneto-optical phenomena are directly or indirectly caused by the splitting energy levels (Zeeman effect) in an external magnetic field\[3\]. Garnets with different dopants show different magnetic and optical properties\[5, 8, 18\] especially Faraday rotation angle, absorption spectra, and saturation field. In this study, by doping gallium into BIG film, we successfully synthesized a new garnet: Bismuth iron gallium garnet (BIGG). BIGG films are more transparent than BIG films, and concurrently keep large Faraday effect.

Iron Borate (FeBO\(_3\)) is another material that is far superior in terms of the figure of merit in the visible spectrum at room temperature to most garnet materials. The interest in iron borate (FeBO\(_3\)) crystal arises from its good transmittance, reasonable Faraday Effect, and low saturation field in the visible range of the spectrum. FeBO\(_3\) crystal was first grown in 1963\[19\], and the applications of magneto-optical properties were discussed in 1970 by Wolfe et al\[20, 21, 22\]. FeBO\(_3\) green platelets we’ve grown show a (001) crystallization orientation. The crystal structure of iron borate is not cubic and has a large natural birefringence for the light propagated along the magnetization\[23-25\]. To eliminate birefringence and increase the transmittance a high energy ball-milling technique was used in this study to synthesize FeBO\(_3\) nanoparticles. The milled
nanoparticles were then hot-pressed to form a new crystal where the orientations of
nanoparticle were randomized and the birefringence was expected to be eliminated. We
also expect that the transmittance of FeBO₃ nanoparticles will be improved because of
the quantum size effect: a decrease in size results in a “blue shift” in the absorption
spectrum. Further investigation and experiments are needed.

Garnet nanoparticles synthesized by coprecipitation and annealing processes such
as Bi₁.₈Y₁.₂Fe₅O₁₂ (BYIG) do not require an expensive substrate and their yield is also
much higher than those obtained with the deposition techniques [26-29]. Similar to BIGG
films where gallium ion was doped to substitute a partial of Iron ions, a new garnet
nanoparticles Bi₁.₈Y₁.₂Fe₄Ga₁O₁₂ (BYIGG) was successfully fabricated in our lab. The
BYIGG powders are more yellowish than BYIG powders.

The optical and magneto-optical properties of one-dimensional photonic crystals
(1D_PCs) of periodic multilayer structures with one or more defects has been studied by
researchers theoretically [17, 30-37] and experimentally [17, 32] in cases where at least one of
the component materials is magnetic. Because of their considerable localization of light,
these media exhibited both enhanced Faraday rotation and transmittance at selected
wavelengths. In the theoretical research part, we focused on the study of an aperiodic
multilayer structure to improve both Faraday rotation and transmittance at the wavelength
of 532nm. The materials used in our analysis were composed of Bi-substituted yttrium–
iron–garnet films and dielectric SiO₂ films. By using a genetic algorithm and a pattern
recognition method, we successfully optimized arbitrary multilayer structures of
magneto-optical materials, which possess higher Faraday rotation and/or transmittance
theoretically.
1.3 Thesis Structure

Iron garnets and rare-earth orthoferrites are two main but different kinds of good magneto-optical materials discussed in this study. In chapter 2, magneto-optical backgrounds of these two materials are described. The Faraday effect and birefringence are included with an emphasis on material properties. In addition, the molecular-orbital energy-level diagram of the crystal structure is described, and the magnetization mechanism and domain structure of the materials are also discussed.

In Chapter 3, the synthesis of our new Bismuth Iron Gallium Garnet thin-films (Bi$_3$Fe$_4$Ga$_4$O$_{12}$, BIGG) by a pulsed laser deposition technique is presented and their properties are compared with these of Bismuth Iron Garnet (BIG) thin-films. The transmittance and Faraday rotation angle of BIGG films are evaluated at the wavelength of 532nm and the magneto-optic figure of merit of BIGG is presented in comparison with BIG films. Further, a switching speed testing on the higher transmittance of our BIGG garnet films is included with a Φ1 mm circular aperture at the wavelength of 532nm.

In chapter 4, the synthesis and testing of Iron Borate crystals are presented, and how their birefringence affects the Faraday rotation is discussed. To eliminate the birefringence and further improve the transmittance, iron borate crystal platelets are ball-milled into nanoparticles, and the milling processes and discussion are included.

Bismuth Yttrium Iron Gallium Garnet (BYIGG) nanoparticles are another new kind of magneto-optical materials we synthesized. The high energy ball-milling and spin-coating techniques produced films with both a higher transmittance and a reasonable Faraday rotation, and these techniques and results are discussed in chapter 5.
In chapter 6, we focus on the design of an RF driving circuit and a miniature coil to overcome the limitation of the driving circuits. The miniature coil was used to generate the required magnetic field of larger than 100 Oe. The response time of the circuit board we designed has reached less than 1ns.

With genetic algorithm and a pattern recognition method, we conducted a theoretical study on how to increase both Faraday rotation and the transmittance with arbitrary multilayer structure, which is discussed in detail in chapter 7. Optimized aperiodic photonic crystal structures of magneto-optical materials are determined, which possess higher Faraday rotation and/or transmittance.

The summary of this study is given in Chapter 8. Future study discussions are also presented in this chapter.

1.4 References


Iron garnets and rare-earth orthoferrites are two different kinds of magneto-optical materials that have been widely studied and their atomic structures, optical and magneto-optical properties are quite different. Iron garnets have a structure of space group Ia3d (#230) with good Faraday rotation but low transmittance in the shorter wavelengths. Rare-earth orthoferrites have good transmittance in the visible region, but they have natural birefringence and their Faraday rotations are small. In this chapter, we will discuss and analyze these properties based on electromagnetic theories in our interested shorter wavelength, 532 nm. In addition, molecular-orbital diagram and magnetization mechanism are also discussed.

The Faraday Effect and birefringence are presented with an emphasis on material properties in section 2.1; the molecular-orbital energy-level diagram of the crystal structure is explained in section 2.2; and the magnetization mechanism and domain structure of material are included in section 2.3.

2.1 Magneto-optical Materials and their Faraday Effect

In 1895, Faraday discovered that when a magnetic field is applied to a block of magneto-optic (MO) material and when a plane wave polarized in a direction parallel to the applied field passes through the MO material, the orientation of the polarization will
rotate a certain degree with respect to the magnetic field applied, which is the famous Faraday Effect \cite{1}.

According to his statement, the rotation angle of the polarization direction $\theta$ is proportional to the magnitude of the parallel magnetic intensity $H$ and the distance $z$ traveled by the light in a medium along the direction of $H$. This relationship can be shown as in Equation 2-1 where $V$ is the Verdet constant, or, the rotation constant per unit path per unit field strength. The Verdet constant $V$ depends upon the properties of the medium, the frequency of the light, and the temperature $T$.

$$\theta = VH_z$$

Eq. 2-1

The Faraday effect is a result of ferromagnetic resonance where the permeability of a material could be represented by a tensor. The resonance causes waves to be decomposed into two circularly polarized rays which propagate at different speeds. The output beam can be considered to re-combine upon emergence from the magneto-optical material. Due to the difference in these two propagation speeds, a net phase shift between these two rays results in a rotation of the linear polarization direction.

### 2.1.1 Magneto-optical Properties of Garnet Materials

Magneto-optical garnet crystals and thin-films are important materials in optical devices. Most garnet Faraday rotators and modulators operate in the infrared range because of their reasonable Faraday rotation and high transmittance in that range. The Faraday rotation of the same garnet material in a shorter wavelength of the visible
spectrum is about 10 to 30 times that in the infrared range \cite{2-4}. However, the absorption is also very high in the visible range, which made it impractical for commercial optical devices. To thoroughly understand the optical properties in the short wavelength of the visible spectrum, a typical bismuth substitute garnet is used to simulate the Faraday rotation and transmittance in this section.

The magneto-optical properties of a given single layer magneto-optical garnet can be derived from the electromagnetic theory, if the relative permittivity tensor and thickness of the magnetic material are given. Two methods, a 4X4 transfer matrix formalism \cite{5-7} and a 4X4 state matrix approach \cite{8-10}, have been developed to theoretically investigate the Faraday effect and transmittance. Following the 4X4 state matrix approach, we will discuss and examine the Faraday effect and transmittance of a typical bismuth substituted yttrium iron garnet (BYIG) material at the wavelength of 532nm in the following.

Suppose linearly polarized TM light (E component parallels to x axis) enters perpendicularly to a film plane at Z=Z0, where the light wave has a form of $e^{-i(kZ-\omega t)}$ with a wave number $k$ and angular frequency $\omega$ as shown in Maxwell’s Equations 2.2.

\[ \nabla \times \vec{E}(\vec{r},t) = i\omega\mu_0\vec{H}(\vec{r},t) \]
\[ \nabla \times \vec{H}(\vec{r},t) = -i\omega\varepsilon_0\varepsilon\vec{E}(\vec{r},t) \]

Eq. 2.2

For a typical BYIG material, the permittivity tensor $\varepsilon$ has additional off-diagonal terms. The nature of Eigen-modes and their corresponding propagation velocities can be derived from Equations 2.2. The tensor is usually expressed through complex permittivity
tensors as in Equation 2-3, where \( \varepsilon_1 \) and \( \varepsilon_2 \) are complex numbers \( \varepsilon_i = \varepsilon_i^r + i\varepsilon_i^i \), and \( \varepsilon_2 = \varepsilon_2^r + i\varepsilon_2^i \).

\[
\tilde{\varepsilon} = \begin{bmatrix}
\varepsilon_1 & i\varepsilon_2 & 0 \\
- i\varepsilon_2 & \varepsilon_1 & 0 \\
0 & 0 & \varepsilon_3 \\
\end{bmatrix}
\]

Eq. 2-3

The tensor elements are related to the refractive index \( n \) and extinction coefficient \( \kappa \) as \( \varepsilon_1^r = n^2 - \kappa^2 \), \( \varepsilon_1^i = 2n\kappa \), \( \varepsilon_2^r = n\Delta n + \kappa\Delta \kappa \), \( \varepsilon_2^i = -n\Delta n + \kappa\Delta \kappa \) where \( n = \left( n_1 + n_2 \right)/2 \), \( \Delta n = n_1 - n_2 \), \( \kappa = \left( \kappa_1 + \kappa_2 \right)/2 \), \( \Delta \kappa = \kappa_1 - \kappa_2 \) for the left (1) and right (2) circularly polarized light. When a magnetic field is applied, the off-diagonal terms in the tensor are proportional to the components of the magnetism. This effect in turn affects the phase of the light field, and the orientation of the polarization will change accordingly.

The light state vector is defined as \( \tau(Z) = \begin{pmatrix}
\varepsilon_x & \varepsilon_y & h_x & h_y \\
\end{pmatrix}^t \)

where \( \varepsilon_0 \) and \( \mu_0 \) are the permittivity and permeability of the vacuum and \( c \) the light vacuum velocity. With the 4X4 state matrix approach, the output state vector can be specified from initial and boundary conditions. Specifically, by using the front and back traveling light with right and left circular polarization in a magnetic media, the state vector can be described by Equation 2.4, where \( k_p = (\omega/c)\sqrt{\varepsilon_p} \), \( k_n = (\omega/c)\sqrt{\varepsilon_n} \) are the wave numbers, \( \varepsilon_p = \varepsilon_1 + \varepsilon_2 \), \( \varepsilon_n = \varepsilon_1 - \varepsilon_2 \) are the right and left circularly polarized permittivities, respectively. A, B, C, and D are the coupling coefficients.
Suppose the thickness of a magnetic dielectric single layer is \( d = Z - Z_0 \), the state vector before entering and after exiting the magnetic dielectric layer can be written as in Equations 2.5, where \( \Phi \) is a 4X4 matrix determined by matrix M and S.

\[
\tau(Z) = A \begin{bmatrix} 1 \\ -i \\ i \sqrt{\epsilon_p} \\ \sqrt{\epsilon_p} \end{bmatrix} e^{ikZ} + B \begin{bmatrix} 1 \\ -i \\ i \sqrt{\epsilon_n} \\ \sqrt{\epsilon_n} \end{bmatrix} e^{-ikZ} + C \begin{bmatrix} i \\ -i \\ i \sqrt{\epsilon_n} \\ \sqrt{\epsilon_n} \end{bmatrix} + D \begin{bmatrix} i \\ i \sqrt{\epsilon_n} \\ \sqrt{\epsilon_n} \end{bmatrix} e^{-ikZ}
\]

\text{Eq. 2.4}

Suppose the thickness of a magnetic dielectric single layer is \( d = Z - Z_0 \), the state vector before entering and after exiting the magnetic dielectric layer can be written as in Equations 2.5, where \( \Phi \) is a 4X4 matrix determined by matrix M and S.

\[
\tau(Z + d) = \Phi \tau(Z)
\]

\text{Eq. 2.5}

Where: \( \Phi = MSM^{-1} \)

\[
M = \begin{bmatrix} 1 & 1 & 1 & 1 \\ -i & -i & i & i \\ \sqrt{\epsilon_p} & i \sqrt{\epsilon_p} & i \sqrt{\epsilon_n} & \sqrt{\epsilon_n} \\ i \sqrt{\epsilon_p} & \sqrt{\epsilon_p} & -i \sqrt{\epsilon_n} & -\sqrt{\epsilon_n} \end{bmatrix}
\]

\[
S = \begin{bmatrix} e^{i\delta_p} \\ e^{-i\delta_p} \\ e^{i\delta_n} \\ e^{-i\delta_n} \end{bmatrix}
\]

The boundary conditions are given in Equations 2.6 considering that the tangential components of the electromagnetic field are continuous at front and back surfaces.
The coupling coefficients $C_1$, $C_2$, $C_3$, and $C_4$ can be solved when substituting boundary conditions of Equations 2.6 into Equations 2.5.

Next, the spectrum of reflectance and transmittance of the light, and the FR can be found from Equation 2.7

$$\tau(z \leq 0) = \begin{bmatrix} 1 \\ 0 \\ 0 \\ 1 \end{bmatrix} e^{ikz} + C_1 \begin{bmatrix} 1 \\ 0 \\ 0 \\ -1 \end{bmatrix} e^{-ikz} + C_2 \begin{bmatrix} 0 \\ 0 \\ -1 \\ 0 \end{bmatrix}$$

$$\tau(z \geq Z) = C_3 \begin{bmatrix} 1 \\ 0 \\ 0 \\ 1 \end{bmatrix} e^{-ik(z-Z)} + C_4 \begin{bmatrix} 0 \\ 1 \\ -1 \\ 0 \end{bmatrix} e^{ik(z-Z)}$$

The coupling coefficients $C_1$, $C_2$, $C_3$, and $C_4$ can be solved when substituting boundary conditions of Equations 2.6 into Equations 2.5.

Figure of Merit (FOM) is defined as in Equation 2.8 where $\alpha_F$ is the Faraday rotation (FR) angle per thickness is degree per micron ($^\circ/\mu$m), $\alpha$ absorption coefficient ($\mu$m$^{-1}$), $d$ the film thickness in $\mu$m, $T$ the total transmittance (0-100%). To avoid the definition confusion, the unit of FOM used is degree ($^\circ$), the same definition as in Kahl [11].
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The off-diagonal components of the tensor depend on the magnetization $M$ of the material, as the applied external magnetic field changes, the magnetization $M$ will change until it reaches saturation. It is assumed here that our garnet is under the saturation field condition, where the tensors are constants.

The diagonal and off-diagonal dielectric elements under saturation field of a typical BYIG film we used in our simulation are listed in Table 2-1 [9].

**Table 2-1: A typical BYIG film’s diagonal and off-diagonal dielectric elements under the saturation field**

- $\varepsilon_1 = 6.09 + 0.05i$
- $\varepsilon_2 = 0.018 + 0.008i$

To give a graphic representation of the optical and magneto-optical properties of garnet materials at the wavelength of 532nm, we programmed Equations 2.3 to 2.8 in Matlab. Figure 2-1 shows the transmittance ($T$), the Faraday rotation angle ($\theta_F$), and the figure of merit (FOM) verses the thicknesses ($d=0.1 \mu m$ to $15 \mu m$) of a typical BYIG garnet at the 532nm wavelength.
It is clearly shown that the transmittance drops and fluctuates with increasing thickness. The Faraday rotation increases but fluctuates slightly with increasing thickness. The figure of merit fluctuates but tends to a fixed value with increasing thickness. Therefore, it can be concluded that the figure of merit of a thin film fluctuates more severely. For example, for the above garnet material, the figure of merit of a 0.7μm thick film could be very different from that of a 1.3μm thick film. Therefore, the thin film’s thickness is another important factor for the magneto-optical device design.

The optical and magneto-optical properties of garnet materials also depend on the wavelength of an incident beam. Figure 2-1 shows the Faraday rotation (FR), the

Figure 2-1: Faraday rotation, Transmittance and Figure of Merit vs. thickness
transmittance (T), and the figure of merit (FOM) verses the wavelength of a typical 1μm BYIG garnet. It is clearly shown that the transmittance drops a little and fluctuates as the wavelength decreases. The Faraday rotation increases but fluctuates slightly with decreasing wavelength. The figure of merit fluctuates but tends to a fixed value with increasing wavelength. Therefore, it is expected that the figure of merit of a thin film fluctuates more severely with wavelength change. Since the transmittance of a 1μm typical BYIG garnet is 99.23% at wavelength of 788nm, the Figure of merit is 45°. However, the figure of merit is not a good measure factor to compare between different wavelengths because the high transmittance in the longer wavelength results much higher Figure of merit \[^{11}\].
Figure 2-3 shows the Faraday rotation (FR), the transmittance (T), and the figure of merit (FOM) verses the wavelength of a typical 1μm, 2μm, 3μm, and 4μm BYIG garnet. We concluded that the Faraday rotation, the transmittance, and the figure of merit depend both on thickness and wavelength.
2.1.2 Faraday Effect and Birefringence in Uniaxial Crystals

For the orthorhombic symmetry crystals[12-15], a large natural birefringence exists for the light propagated along the magnetization direction. FeBO$_3$ crystal is one of orthoferrites of our interest and has also a good transmittance and a reasonable intrinsic Faraday rotation. This section gives a theoretical explanation why the birefringence could reduce the maximum obtainable rotation in the orthorhombic crystals.

Figure 2-3: Faraday rotation, Transmittance and Figure of Merit vs. wavelength $\lambda$ and thicknesses

2.1.2 Faraday Effect and Birefringence in Uniaxial Crystals

For the orthorhombic symmetry crystals[12-15], a large natural birefringence exists for the light propagated along the magnetization direction. FeBO$_3$ crystal is one of orthoferrites of our interest and has also a good transmittance and a reasonable intrinsic Faraday rotation. This section gives a theoretical explanation why the birefringence could reduce the maximum obtainable rotation in the orthorhombic crystals.
Suppose an anisotropic crystal’s a, b, and c axes are parallel to the Cartesian coordinate system x, y, and z axis, respectively. When a net magnetization is along $\pm z$, and it is assumed the material has no absorption, the permittivity tensor is shown in Equation 2.9.

$$\varepsilon = \begin{bmatrix} \varepsilon_1 & i\gamma & 0 \\ -i\gamma & \varepsilon_2 & 0 \\ 0 & 0 & \varepsilon_3 \end{bmatrix}$$ \hspace{1cm} \text{Eq. 2.9}

Using Maxwell’s Equations 2.2 and a plane wave propagating along the z axis of the crystal, Equation 2.10 can be derived.

$$\begin{pmatrix} \varepsilon_1 - \frac{k^2}{\omega^2 \mu} & i\gamma \\ -i\gamma & \varepsilon_2 - \frac{k^2}{\omega^2 \mu} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \end{pmatrix} = 0$$ \hspace{1cm} \text{Eq. 2.10}

To find the nontrivial solution of the algebraic equations, the determinant needs to vanish. The two Eigen-value $k_\pm$ are found in Equation 2.11.

$$\left(k_\pm\right)^2 = \frac{1}{2} \omega^2 \mu \left(\varepsilon_1 + \varepsilon_2\right) \pm \left[\left(\varepsilon_1 - \varepsilon_2\right)^2 + 4\gamma^2\right]^{1/2}$$ \hspace{1cm} \text{Eq. 2.11}

The Eigen-vectors of the two characteristic modes are shown as in Equation 2.12, where $\alpha = 2\gamma / \left[2\varepsilon_1 - \left(\varepsilon_1 + \varepsilon_2\right) \pm \left[\left(\varepsilon_1 - \varepsilon_2\right)^2 + 4\gamma^2\right]^{1/2}\right]$.

$$\begin{pmatrix} E_x' \\ E_y' \end{pmatrix} = A \begin{pmatrix} 1 \\ -i/\alpha \end{pmatrix} \exp[i(\omega t - k_+ z)]$$ \hspace{1cm} \text{Eq. 2.12}

$$\begin{pmatrix} E_x'' \\ E_y'' \end{pmatrix} = B \begin{pmatrix} 1 \\ i\alpha \end{pmatrix} \exp[i(\omega t - k_- z)]$$
Since the characteristic modes are not convenient to work with, we will transform Equation 2.12 into a Cartesian coordinate system and find the E-field intensities between $z=0$ to $l$ as expressed in Equation 2.13, where $\kappa = \delta z$, $\delta = k_+ - k_-$, 
\[ \cos \tau = \frac{(1 - \alpha^2)}{(1 + \alpha^2)}, \text{ and } \sin \tau = 2\alpha / (1 + \alpha^2). \]

\[
\begin{pmatrix}
E_x \\
E_y
\end{pmatrix}
\bigg|_{z=d} = \begin{pmatrix}
\cos(\kappa/2) - i \cos \tau \sin(\kappa/2) & -\sin \tau \sin(\kappa/2) \\
\sin \tau \sin(\kappa/2) & \cos(\kappa/2) + i \cos \tau \sin(\kappa/2)
\end{pmatrix}
\begin{pmatrix}
E_x \\
E_y
\end{pmatrix}
\bigg|_{z=0}
\]

If a linear polarized beam enters perpendicularly on the orthorhombic crystal, Equations 2.14 is obtained at $z>0$. Therefore, the polarization components $E_x$ and $E_y$ can be obtained if the wavelength $\lambda$, thickness $d$, and all components of the permittivity tensor $\varepsilon$ are known.

\[
E_x = \cos(\kappa/2) - i \cos \tau \sin(\kappa/2) \tag{Eq. 2.14}
\]
\[
E_y = \sin \tau \sin(\kappa/2)
\]

Two cases are clearly shown, when $\sin \tau = 1$, $E_x = \cos(\delta z/2)$ and $E_y = \sin(\delta z/2)$, Equations 2.14 correspond to the pure Faraday effect. When $\sin \tau = 0$, $E_x = \exp(-i \delta z/2)$ and $E_y = 0$, Equations 2.14 correspond to the pure birefringence. The maximum value of $E_y$ is $\sin \tau$, the relative strength of the Faraday rotation to the birefringence $\delta$, the total relative phase per unit length. These two parameters, $\sin \tau$ and $\delta$, could be used to describe the orthoferrite material’s Faraday rotation and birefringence.

The polarization ellipse can be expressed in terms of two angular parameters: the orientation angle $\psi$ and the ellipticity angle $\chi$ as shown in Equation 2.15.
Since the $\sin \tau$ is small for the orthoferrites [15], Equation 2.15 can be simplified as shown in Equation 2.16.

$$\psi = \sin \tau \sin \kappa / 2$$  \hspace{1cm} \text{Eq. 2.16}$$

$$\chi = \sin 2\tau \sin^2 (\kappa/2) / 2$$

It is clearly shown that the orientation angle $\psi$ and the ellipticity angle $\chi$ can not be large. Although the intrinsic Faraday rotation can be large (0.23°/μm for FeBO$_3$), the maximum rotation for orthoferrite single crystals for propagating along the c-axis is limited by the sinusoidal functions. Therefore, an orthoferrite material with birefringence will not be capable of rotating the plane of polarization by 90° as it would when no birefringence exists.

### 2.2 Molecular-Orbital Energy-Level Diagram

The Zeeman effect is the splitting of a spectral line into several components in the presence of a magnetic field [16]. In most atoms, there exist several electronic configurations that have the same energy, so the transitions between different pairs of configurations correspond to a single line. However, with an external magnetic field applied, the degenerated energy level is broken. The result is that, where there are several configurations with the same energy, there are different energies, which give rise to several very close spectral lines.
The Molecular-Orbital Energy-Level Diagram approach could clearly explain the relations between the rotation angle and the incident beam spectra, and the corresponding formula fit the Faraday rotation spectra for the visible beam successfully\cite{17-19}.

The rotation and ellipticity spectra of Bismuth doped YIG crystals can be interpreted with diamagnetic functions described by the off-diagonal permittivity tensor elements $\epsilon_i$. The Faraday rotation Verdet constant can be rewritten in Equation 2-17, according to the molecular-orbital energy-level diagram of Allen and Dionne\cite{17-19}, where the off-diagonal permittivity tensor elements $\epsilon_i$ can be derived from Equation 2-18.

$$\theta_F = \frac{\omega}{2nc} \epsilon_i$$  \hspace{1cm} (Eq. 2-17)  
$$\epsilon_i = \omega_p^2 \sum \pm \frac{\omega}{2\omega_{0z}} \frac{\omega(\omega_{0z}^2 - \omega^2 - \Gamma^2) + i\Gamma(\omega_{0z}^2 + \omega^2 + \Gamma^2)}{\left(\omega_{0z}^2 - \omega^2 + \Gamma^2\right)^2 + 4\omega^2\Gamma^2}$$  \hspace{1cm} (Eq. 2-18)

In Equation 2-18, the sum is over the right- and left-hand circular polarized light.

The $\omega_{0z} = \omega_0 \pm \Delta$, represented as right- and left-hand circular polarized light respectively, $\omega_p^2 = 4\pi Ne^2 / m$, whereas N is the active ion density, e and m are the electron charge and mass, $f_\pm$ are the respective oscillator strengths, and $\omega_0$ and $\Gamma$ are the frequency and the half-line width of the transition between the ground $|g\rangle$ and the excited state denoted by $|e_\pm\rangle$ that is splitted by $2\Delta$.

The molecular-orbital energy-level diagram can successfully fit the Faraday rotation spectra in the visible range of the spectrum. Figure 2-4 shows a typical
simulation result for BYIG garnet. The largest rotation angle found at garnet films corresponds to the wavelength 515nm.

Dionne\cite{18} proposed a molecular-orbital energy-level diagram of Bi$_3$Fe$_5$O$_{12}$ (BIG). Its molecular-orbital energy-level diagram was shown in the Figure 2-5 where the oxygen interactions and all antibonding states have been omitted to reduce confusion. The large Faraday effects of BIG arise from the cooperative action of Fe$^{3+}$ ions with degenerate orbital terms that are split further by covalent interaction with Bi$^{3+}$.
2.3 Magnetization Mechanism

In magnetic materials, domains are small regions within which all the magnetic dipoles are aligned parallel to each other\[^{16, 20}\]. Domains form in magnetic materials as a result of minimizing the total energy. The boundaries between adjacent domains in bulk magnetic materials are called domain walls. When a magnetic material is in its demagnetized state, the magnetization vectors in different domains have different orientations, but the total magnetization averages to zero.

When a magnetic field is applied to a ferromagnetic material, the magnetization tends to align along a certain preferred crystallographic direction. The preferred direction
is called the 'easy' axis, since it is easiest to magnetize a demagnetized sample to saturation if the external field is applied along this preferred direction.

After the applied magnetic field is removed, the dipoles rotate back to their easy axis of magnetization, and the net magnetic moment along the field direction decreases. Because the demagnetization process is driven by the demagnetizing field, rather than an applied external field, the demagnetizing field is not strong enough to overcome the energy barriers encountered when the domain walls intersect crystal imperfections. As a result, some magnetization remains in the sample even when the field is removed completely, which is shown as a hysteresis curve when we measure the magnetization vs. applied field. The coercive field is defined to be the additional field to reduce the magnetization to zero in the reverse direction.

The exchange energy prefers a wide wall while the magnetocrystalline anisotropy energy prefers a narrow wall. The width of a domain wall depends on the balance between the exchange energy and the magnetocrystalline anisotropy energy. When a crystal has large magnetocrystalline anisotropy, large single-domain particles can be formed because the domain wall energy is relatively large. However, if the saturation magnetization is large, then the magnetostatic energy is large too. In this case, a domain wall formation is favorable, so a large single domain can not be formed.

Figure 2-6 shows a magnetization mechanism in a single-domain magnetic material. Before an external field is applied, the magnetization of a single-domain particle lies along an easy direction as in Figure 2-6 (1). When an external field is applied in the opposite direction, the particle is unable to respond by domain wall motion, and instead the magnetization must rotate through the hard direction as in Figure 2-6 (2) to the new
easy direction as in Figure 2-6 (3). The anisotropy forces which hold the magnetization in an easy direction are strong, and so the coercivity is large. When the magnetic field is applied along an easy direction, the square hysteresis loop is formed as in Figure 2-6 (4). Two bi-polar stable states of opposite magnetization in Figure 2-6 (4) are desirable for magnetic media storing applications.

The speed of magnetization reversal in the configuration in Figure 2-6 can be accelerated by a precessional motion technique [21, 22]. To do this, another static magnetic field $H_0$ is needed to apply to the hard axis direction. The $H_0$ field cancels the anisotropy forces holding the magnetization in the easy direction, and accelerates the rotation of the magnetization through the hard direction. Therefore, the switching speed of this configuration with a precessional technique could be greatly improved.

If the field is applied along a hard direction as shown in Figure 2-7 (1), there is initially no component of magnetization along the field direction. The field rotates the magnetization into the field direction as in Figure 2-7 (2), and it rotates back into the easy
direction as soon as the field is removed as in Figure 2-7 (3). As a result, there is little hysteresis and the B-H curve is approximately linear as shown in Figure 2-7 (4). Therefore, crystals with a very small coercivity are prone to fast response to external magnetic field, and the particles in the crystal will align with their easy axes perpendicular to the applied field direction.

As discussed above, the speed for the magnetization reversal is closely related to the magnetization direction of the domain dipoles and configurations. Therefore, the configuration and domain structure of a magnetic material are the important factors to be considered in our ultrafast switch application. Two configurations shown in Figures 2-6 and 2-7 have been tested in our experiments. The most used configurations in our testing are the same as Figures 2-6 where the easy plane is the in-plane of the material.
2.4 References


Chapter 3

Synthesis of New Bismuth Iron Gallium Garnet Thin-films and their Optical and Magneto-optical Properties

3.1 Introduction

Bismuth Iron Garnet (Bi$_3$)$_5$Fe$_3$O$_{12}$, BIG) films have been widely studied in the past two decades as a promising material for magneto-optical applications due to their giant Faraday rotation as high as 7 $^\circ/\mu$m at 633 nm wavelength$^{[1-5]}$. They are promising for various applications such as isolators, modulators, and switches in communication systems. Since the BIG film is not thermodynamically stable and cannot be synthesized by a liquid phase epitaxial method$^6$, a pulsed laser deposition (PLD) technique was used to synthesize BIG films. This technique offers exact transfer of stoichiometry with a surface quality of films comparable with those of physical vapor depositions$^2$.

In our effort to search for a high Faraday effect and high transmittance host material for applications in ultrafast switching in the visible range of the spectrum, garnet thin-films were chosen for study. Although a BIG thin-film has a high Faraday effect, its transmittance is very low at wavelengths shorter than 550 nm$^{[6-9]}$. New garnet thin-films were synthesized by substituting the ions in different sites of the BIG garnet.

The chemical formula of an ideal BIG film is {Bi$_3$}[Fe$_2$(Fe$_3$)O$_{12}$ where Bi$^{3+}$ ions occupy the dodecahedral sites, two of the Fe$^{3+}$ ions occupy octahedral sites and the remaining three Fe$^{3+}$ ions reside in tetrahedral sites. Lanthanum or other rare-earth ions can replace bismuth ions in the dodecahedral sites, while aluminum and gallium have
strong preference for substituting into the tetrahedral sites, and indium and scandium prefer the octahedral sites \[^{[10]}\].

BIG’s giant Faraday effect is attributed to the Bi\(^{3+}\) ions perturbing and enhancing the multiple splitting of the excited band of the Fe\(^{3+}\) ions\[^{[11-13]}\]. It is known from films grown by a liquid phase epitaxy method that the Faraday rotation increases linearly at a rate of 2.06 \(°/\mu\text{m}\) at 633 nm\[^{[6]}\] and 5.3 \(°/\mu\text{m}\) at 539 nm\[^{[9]}\] wavelength per bismuth atom per formula unit. Therefore, to keep its high Faraday effect, bismuth ions should not be disturbed.

In our effort, a new garnet was synthesized by doping gallium ions to replace partial iron ions in the tetrahedral sites. The gallium doped in the tetrahedral sites may reduce the Faraday effect by about 1.4 \(°/\mu\text{m}\) at 539 nm wavelength per gallium atom per formula unit\[^{[9]}\]. However, considering the high transmittance of gallium garnets such as terbium gallium garnet, the figure of merit may improve greatly. In this study, Bi\(_3\)Fe\(_4\)Ga\(_1\)O\(_{12}\) (BIGG) film was successfully grown by a PLD technique and characterized with X-ray diffraction, X-ray rocking curve analysis, Energy Dispersive X-Ray Spectroscopy, atomic force microscopy and magnetization hysteresis techniques. Its optical and magneto-optical properties were evaluated at the wavelength of 532nm with a Nd:YAG laser and the magneto-optical figure of merit of BIGG films was presented in comparison with BIG films. Also we performed speed testing with a \(\Phi 1\) mm circular aperture at 532 nm based on the higher transmittance of our BIGG garnet films. A miniature coil was sputtered and etched on top of garnet film for the driving magnetic field. Its switching response time was tested to have reached 2.4 ns.
3.2 Film Synthesis

BIG and BIGG have a garnet structure of space group Ia3d (#230). Figure 3-1 shows the structure of our new BIGG material.

![Figure 3-1: Ia3d (230) structure of a BIGG crystal](image)

In the pulsed laser deposition process, a Lambda Physik LPX300 KrF excimer laser was used in BIGG and BIG film deposition. The laser wavelength was 248nm, the pulsed energy density was about 2–3J/cm², and the repetition rate used was 20Hz.

Figure 3-2 shows a schematic illustration of Pulsed Laser Deposition.
The BIGG target was made of bismuth oxide, iron oxide, and gallium oxide powders. The stoichiometric Bi:Fe:Ga molar ratio of the target was chosen to be 3:4:1, which was required for the Bi$_3$Fe$_4$Ga$_1$O$_{12}$ phase. The powders were ball-milled for 48 hours to ensure microscopic homogeneity. Then it was cold-pressed at 300kg/cm$^2$ and sintered at 825°C for 4 hours. Taking into consideration the volatility of bismuth during high temperature sintering and deposition processes, another target with 20% extra stoichiometric ratio of bismuth had also been prepared. X-Ray Diffraction (XRD) analysis confirms that the targets had several phases: Bi$_2$Ga$_4$O$_9$, Bi$_{25}$FeO$_{40}$, Bi$_{24}$Ga$_2$O$_{39}$, and Bi$_{25}$GaO$_{40}$; the BiFeO$_3$ phase is not found in the BIGG target as shown in Figure 3-3.
Gd$_3$Ga$_5$O$_{12}$ (GGG) and Gd$_3$(ScGa)$_5$O$_{12}$ (GSGG) crystals were used as the substrates. During the deposition, a substrate was heated and kept at a growth temperature between 500° and 585 °C. The distance between the substrate and the target was set at about 5 cm and the typical deposition time was around 40 minutes. The oxygen partial pressure was controlled from 5 to 20 mTorr. The deposition rate was as high as about 3 $\mu$m/hr.

The thicknesses of BIG and BIGG films were calculated by the interference fringe pattern of their transmission spectrum$^{[14]}$ and verified by a profilometer. An example is illustrated in Appendix 1 to calculate the thickness of a thin-film on a thicker substrate. To make a sharp step on the film, a small part of the substrate was either masked with a metal strip before deposition or alternatively, a small part of the film was etched away.
with acid as shown in the inset of Figure 3-11. The calibrated deposition thickness per pulse was used to estimate the thickness before the deposition.

### 3.3 Film Characterization and Analysis

X-Ray Diffraction (XRD), Rocking curves and lattice constant of BIGG are reported in section 3.3.1. Energy Dispersive X-Ray microanalysis (EDX) results of the films is shown in section 3.3.2. The hysteresis Curves are measured in section 3.3.3. Grains sizes of BIGG and BIG are measured in section 3.3.4. The Faraday rotation, transmittance and the figure of merit of BIG and BIGG films are measured and the results are analyzed in section 3.3.5. The result of switching response of the BIGG film is shown in section 3.3.6.

#### 3.3.1 X-ray diffraction, Rocking Curves and Lattice Constant

Waves, such as X-rays, scatter and diffract when they struke a set of regularly spaced lattices whose dimensions are of the order of the wavelength. The regularly spaced atoms in a crystalline material are effective in scattering waves with wavelength comparable to atomic dimensions. X-ray diffraction [15, 16] has become a powerful tool for studying the arrangement of atoms in a crystal.
In 1912, Bragg used X-rays to study the structure of crystalline solids and proposed a simple model to explain the occurrence of interference maxima in X-rays scattered by a crystal. A beam of parallel X-rays incident at an angle $\theta$ measured from the plane is thus reflected at $\theta$ from the plane as shown in Figure 3-4. Reflections from successive planes of spacing $d$ travel along paths which differ in length by $2dsin\theta$. Therefore, reflected waves interfere with each other. This is the famous Bragg condition for constructive interference as shown in Equation 3.1 where $d$ is the spacing between the adjacent planes in the parallel set, $\theta$ is the Bragg angle, $\lambda$ is the wavelength of the X-rays.

$$2d \sin \theta = n\lambda \tag{Eq. 3.1}$$

For a cubic crystal, the distance (d-spacing) between planes (hkl) is shown in Equation 3.2, where “a” is the lattice constant.
A Scintag X-ray powder diffractometer with Cu tube was used to study the films’ crystalline structure. The wavelengths of Cu Kα- and Kβ- lines are 1.54059Å and 1.54442Å, respectively. Based on the garnet structures with the space group symmetry of Ia3d (body centered cubic), the lattice constants of the substrates and films were calculated. The lattice constant of the GGG substrate was found to be 12.38 Å, and that of the GSGG substrate was 12.48 Å. A lattice constant of 12.63Å for the BIG film was confirmed and was consistent with most literature [1, 3, 5, 17-20], while that of our BIGG films varied from 12.58Å to 12.63Å depending on the amounts of gallium substitution. The lattice constant of Bi₃Fe₄Ga₁O₁₂ films had a value of 12.61±0.01Å. Figure 3-5 shows a typical XRD result of a BIGG film on a GGG(001) substrate.
The lattice mismatch between a BIGG film and a GGG substrate can be seen from XRD. The smaller the lattice mismatch, the lower the coercive force between film and substrate, the smaller the saturation field for the magneto-optic switching material, the faster the switching speed. The lattice mismatch between BIGG and GSGG is smaller than that between BIGG and GGG.

The Bragg reflection condition describes an ideal situation. In reality, the energy E is deflected with certain probability in a certain angular range near the Bragg angle $\theta$. The Rocking Curve is used to characterize the peak reflectivity, the full width at half maximum (FWHM) and the position of the RC maximum. Usually we use rocking curves to verify the epitaxial quality of a single crystal.

Figure 3-5: X-Ray Diffraction of a BIGG film on a GGG (001) substrate
Rocking curves of (004) reflections of a BIGG film in comparison with GGG (001) substrate are shown in Figure 3-6. The full width at half maximum (FWHM) was 0.26° for BIGG films and 0.02° for a GGG substrate respectively. It tells us that the BIGG film is of good epitaxial quality, but its epitaxial quality is not as good as that of its GGG substrate.

---

**Figure 3-6**: Rocking curves of (004) reflections of a BIGG film (solid blue line) and a GGG (001) substrate (dashed red line)

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**3.3.2 Energy Dispersive X-Ray Microanalysis**

EDX can be used for identifying the elemental composition of the films. In this experiment, EDX confirmed that BIG film contains the proper molar ratio of bismuth and iron ions while BIGG film has the proper molar ratio of bismuth, iron and gallium ions.
As we discussed in section 3.2, two kinds of targets have been made: one has a normal bismuth molar ratio, and the other, a 20% higher molar ratio of bismuth before sintering. The XRD of films shows that the bismuth is deficient in the films that were deposited with the normal molar ratio targets, while bismuth is close to nominal value in the films that were sputtered with the enriched bismuth target. These bismuth-enriched films have close compositions to their formula ones as shown in Table 3-1. The accuracy of the EDX results is ±15%.

Table 3-1: Figure of merit of different garnet films

<table>
<thead>
<tr>
<th>Short names</th>
<th>Nominal formula</th>
<th>EDX results (±10%)</th>
<th>Faraday angle</th>
<th>Absorp.coeff. $\alpha$ (10$^3$ cm$^{-1}$)</th>
<th>Figure of merit (°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BIG</td>
<td>Bi$_3$Fe$<em>5$O$</em>{12}$</td>
<td>Bi$_{2.7}$Fe$<em>5$O$</em>{12}$</td>
<td>16.2±0.8°</td>
<td>38.0±4.0</td>
<td>9.0±0.5</td>
</tr>
<tr>
<td>BIGG</td>
<td>Bi$_3$Fe$_4$Ga$<em>1$O$</em>{12}$</td>
<td>Bi$<em>{2.8}$Fe$</em>{4.1}$Ga$<em>{0.9}$O$</em>{12}$</td>
<td>15.6±0.6°</td>
<td>18.5±2.5</td>
<td>16.5±1.0</td>
</tr>
</tbody>
</table>

3.3.3 Hysteresis Curve

Hysteresis curves of BIGG films on GSGG (001) and GGG (111) substrates were measured with a Quantum Design PPMS instrument at room temperature in two geometries: when a bias magnetic field was normal to the film plane and when parallel to the film plane. The deposition conditions of the films are listed in Table 3-2.

Table 3-2: BIGG( Bi$_3$Fe$_4$Ga$_1$O$_{12}$) films used for hysteresis analysis

<table>
<thead>
<tr>
<th>#</th>
<th>substrate</th>
<th>orientation</th>
<th>date</th>
<th>Temp</th>
</tr>
</thead>
<tbody>
<tr>
<td>#0041</td>
<td>GGG</td>
<td>111</td>
<td>03/16/06</td>
<td>560</td>
</tr>
<tr>
<td>#0094</td>
<td>GSGG</td>
<td>001</td>
<td>07/07/06</td>
<td>570</td>
</tr>
</tbody>
</table>
The paramagnetic response of the substrate is responsible for the constant slope of total hysteresis measured. Therefore, the paramagnetic component needs be removed to get the magnetization curve of the film. To do this, the film can be etched away from the substrate, which is then measured alone. The slope can then be determined and subtracted from the total H-M curve. However, a 0.5 mm substrate is much thicker than a 1 μm film and the momentum of gadolinium in GGG substrate is also larger, therefore, the magnetization component of the substrate was about 10X higher than that of the film. The subtraction error between 2 large signals may result in a large propagating error. Therefore, we used a fitting method to extract the weak film signal from the total magnetization data without measuring the substrate’s magnetization curve alone. It was assumed that all paramagnetic components came from the substrate and was fitted into a straight line through the coordinate origin. The film’s magnetization curve was then obtained by subtracting the fitting line. The hysteresis M-H curves of BIGG films on GSGG (001) substrates are shown in Figure 3-7.
The hysteresis M-H curves of BIGG films on GGG (111) are shown in Figure 3-8

Figure 3-7: Magnetization M vs H curves of BIGG films in a magnetic field parallel (dashed) and perpendicular (solid) to the film plane on a GSGG(001) substrate

Figure 3-8: Magnetization M vs H curves of BIGG films in a magnetic field parallel (dashed) and perpendicular (solid) to the film plane on a GGG(111) substrate
3.3.4 Grain Size Measurement

Figure 3-9 shows the 3D grain size images of a typical BIG and BIGG film with an atomic force microscope. The Z-axis was scaled 10X larger than X-Y plane. The average grain size in BIG film was between 63 nm and 98 nm, and the average grain sizes in BIGG film was between 82 nm and 200 nm. This may indicate that there was some kind of combination or reaction of the grains in BIGG film via gallium substitution. The lower coercive force in the BIGG film possibly attributed to the magnetic anisotropy changes on account of the substitution of Ga by Fe.
Faraday rotation experiment was done in our lab. For the saturation field testing, a coil was wound on a Φ2.5”X5” cylindrical steel holder with a Φ0.7” hole to contain the sample. The generated magnetic field was up to 2500 Oe. After several designs, the coil met our saturation requirement but heat is generated quickly. Without a cooling system, the power was turned on-and-off quickly during testing to avoid heat accumulation. A small Teflon holder was used to hold the sample and isolate heat transferring from the

Figure 3-9: AFM images of BIG and BIGG films. The Z-axis was scaled 10X larger than X-Y plane
coil. It worked well though the accuracy was relatively low (±15%) at small magnetic field. This coil had a very big inductance and is not used for the switching testing (where a miniature coil was designed separately).

The saturation field is one of the important factors that affect the switching speed of the film. Typically, the saturation field Hs was set at about 1200(Oe) for a BIG film on a GGG substrate, about 400(Oe) for a BIGG on a GSGG substrate. Figure 3-10 shows a typical Faraday rotation test result and the saturation field of a 1.8μm BIGG (100) film on a GSGG substrate under a high magnetic field. The data is collected manually. The orientation of the substrate also slightly affects the saturation field, the (111) film has a saturation field of about 50(Oe) smaller that that of (100) substrates. In our high speed switching testing, it was found the response of the film made on a GSGG substrate was much faster than that on a GGG substrate. The switching response of a BIGG film was slightly better than that of a BIG film because the former had a smaller lattice mismatch.
The thicknesses of BIG and BIGG films were estimated by the interference fringe pattern of the transmission spectrum and later confirmed by an atomic force microscope (AFM) measurement. The transmittance spectra of films were measured by a lambda spectrometer relative to a GGG substrate.

Figure 3-11 shows the transmittance curves which are normalized relative to 1\(\mu\)m thick film. The spectrum curve of our BIG film was consistent with that of Kahl’s measurement\[^6, 17, 21, 22\]. It is clear that the BIGG film was more transparent than the BIG film from the inside pictures that show the colors of ~1\(\mu\)m BIG and ~1.6\(\mu\)m BIGG films. The bandwidth of the BIGG’s transmittance spectrum has been extended and its left edge has been shifted about 50nm to the shorter wavelengths relative to that of the BIG.
According to the crystal field split energy diagram by Scott, the transmittance spectrum depends on the split energy levels of ions in the octahedral and tetrahedral sites. The oscillator strength of the transitions between 333nm and 1000nm depends on the concentration of iron in both sites. Since the doping of gallium lowers the concentration level of iron in tetrahedral sites, the oscillator strength of the transitions in tetrahedral sites has been lowered. This explanation was confirmed by the shifting of the BIGG’s absorption spectrum to the shorter wavelength about 50nm.

Figure 3-11: Transmittance spectrum of BIG and BIGG films with inset: color pictures of BIG and BIGG films (BIG was partially etched for thickness measurement)
To compare and evaluate the whole performance of the films with different thicknesses and different Faraday rotation angles, a figure of merit is used as in Equation 3-3 where FOM is the figure of merit, $\theta_F$ Faraday rotation angle per thickness, $d$ is the thickness of the film, $\theta_F \cdot d$ the total measured Faraday rotation and T% the normalized transmittance of the garnet film. To avoid the definition confusion, the unit of FOM used is degree (°), the same definition as in Kahl [6].

$$FOM = \frac{2|\theta_F \cdot d|}{-\ln(T\%)}$$

Eq. 3-3

This formula can be simplified as $FOM = 2|\theta_F|/\alpha$, where $\alpha$ is an absorption coefficient of the film. Therefore, the figure of merit is not related to the thickness of the film.

Faraday effect experiments were tested with a Nd:YAG 532nm laser beam. Table 3-1 shows the Faraday rotation angle, the absorption coefficient of the film, and the calculated figure of merit for the BIG and BIGG garnet films. Their atomic weight ratio was checked by EDS. Table 3-1 shows the statistical results with the figure of merit of the films obtained so far. Some films have either slightly better Faraday rotation (about $17^\circ/\mu m$ at 532nm) with a lower transmittance or better transmittance with lower Faraday rotation. Typically, the figure of merit of the BIGG films is about 1.8 times higher than that of the BIG film at 532nm. Therefore, the performance of the BIGG film is much better than that of BIG film.
3.3.6 Switching Response

Figure 3-12 shows the schematic setup of the switching response experiment of our magneto-optic materials.

The real setup of switching response experiment is shown in Figure 3-13, while the RF circuit design will be discussed in Chapter 6.
Based on our BIGG garnet films, a switching speed test was performed. The circular aperture of the switching sample was around $\Phi 1\ mm$. A Nd:YAG 532 nm laser was used as the light source. A miniature gold coil was designed, sputtered and etched on the top of a BIGG film. A RF circuit board was carefully designed for high speed response. A 1ns electric current pulse was sent to the switching magnetic coil to generate a switching magnetic field around 100 (Oe) on the magnetic thin-film. Figure 3-14 shows that the measured switching rising time was about 2.41 ns. captured from Tektronix TDS 784C oscilloscope (1 Ghz, 4 Gs/s).
To our knowledge, this is the fastest MO switching speed achieved with a $\Phi 1$ mm circular aperture. Note that this 2.4 ns switching speed (rise time) is mainly limited by the optical detection system and the driving electronics. Also, the signal decay time (fall time, about 10 ns) could be either reduced (by applying an opposite switching magnetic field) or extended (by leveraging the latching capability of magnetic materials) depending on specific application requirements.

3.4 Conclusion

$\text{Bi}_3\text{Fe}_4\text{Ga}_1\text{O}_{12}$ (BIGG) films have been synthesized on $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ and $\text{Gd}_3(\text{ScGa})_5\text{O}_{12}$ substrates by a pulsed laser deposition technique. X-ray diffraction and rocking curve analyses confirm the epitaxial quality of the films with a lattice constant

![Speed pulse response curve of our BIGG film from the oscilloscope](image)
close to 12.61±0.01 Å. Compared with that of a bismuth iron garnet Bi$_3$Fe$_5$O$_{12}$ (BIG) film, the bandwidth of the Bi$_3$Fe$_4$Ga$_1$O$_{12}$ transmittance (T) spectrum was extended and its left edge shifted down in wavelength by approximately 50 nm. This enables its Faraday rotation $\theta_F$ and switching speed to be tested at a wavelength of 532 nm. While Bi$_3$Fe$_4$Ga$_1$O$_{12}$ film had a Faraday rotation angle slightly less than that of BIG film, its magneto-optical figure of merit, $2|\theta_F|d/-\ln(T\%)$, has been greatly improved to a typical value about 1.8 times larger than that of the BIG film. Further, the high speed pulse response time of the Bi$_3$Fe$_4$Ga$_1$O$_{12}$ film has reached 2.4 ns with a Φ1 mm circular aperture at the wavelength of 532 nm.

### 3.5 References


Iron Borate (FeBO₃) is another material that is far superior in terms of the figure of merit (the ratio of the Faraday rotation angle to the absorption coefficient) in the visible spectrum at room temperature to most magneto-optical materials. In this study, highly perfect iron borate platelet-like single crystals were grown by a flux sintering technique and used for our high speed switch testing. The single crystals were tested with X-Ray diffraction and it was shown that our crystal has a new crystal orientation (001). Our switching testing experiments showed that the speed of the switch using FeBO₃ platelets has reached 1.2ns with a Φ1 mm circular aperture at the wavelength of 532nm.

The switching speed of FeBO₃ is high but Faraday rotation cannot reach more than 5° because the light propagating along the magnetization direction undergoes birefringence phase retardation. The birefringence retardation causes the velocity difference of the incident beam’s 2 E-field components. The interference of birefringence on Faraday rotation reduced the maximum obtainable rotation. Therefore, random orientated particles could balance the velocity components and then overcome the limitation of Faraday rotation by birefringence [1]. In our study, a high energy ball-milling technique was used to synthesize FeBO₃ nanoparticles. Our simulation in section 4.3.1 shows the nanoparticles could eliminate the birefringence, and concurrently keep the Faraday rotation. After milling and centrifuging, the FeBO₃ nanoparticles’ sizes...
reached down to 30nm so far. The nanoparticles are to be hot-pressed and re-formed into an orientation-randomized new crystal. It is expected that the nanoparticles approach will overcome the birefringence, improve the transmittance, and concurrently keep the same switching speed.

4.1 Introduction

The interest in iron borate crystals mainly arises from their good transmittance in the visible range of the spectrum, their reasonable Faraday Effect, and their low saturation field \([2-4]\). Besides, their small in-plane anisotropy, large out-of-plane anisotropy and small \(4\pi M_s\) makes them favorable in isolators, electronically tunable filters, wide-band optical modulators and deflectors \([2, 5, 6]\).

Iron borate has a calcite structure (space group \(R\overline{3}C\)) that can be described in terms of hexagonal or rhombohedral axes \([2-4, 7, 8]\). The crystals are dominated by close-packed array of \(\text{Fe}^{3+}\) cations and \((\text{BO}_3)^{-3}\) anions layered upon each other. With CCDC’s Mercury software \([9]\), the crystal structure of \(\text{FeBO}_3\) is shown in Figure 4-1. Note that the red balls in the figure represent oxygen ions, while the tan and blue balls are boron and iron ions, respectively.
Ferric borate FeBO$_3$ was first fabricated in 1963 by Bernal et al\cite{10}. Using a flux system containing Bi$_2$O$_3$ as well as B$_2$O$_3$ and Fe$_2$O$_3$, the authors grew what they described as yellowish-green, well-formed hexagonal platelets. In 1968 a powder technique was used to prepare amorphous FeBO$_3$\cite{2}. Another flux growth technique with B$_2$O$_3$-Fe$_2$O$_3$-PbO/PbF$_2$ was used to grow highly perfect single crystal platelets of FeBO$_3$ in 1985 \cite{3}. Bulk FeBO$_3$ was prepared from the vapor phase in evacuated quartz ampoules via a chemical transport reaction with chlorine and a gas phase reaction of the binary oxides Fe$_2$O$_3$ and B$_2$O$_3$ using HCl as transporting agent\cite{11}. A direct sintering and quenching technique was also used to grow single crystal in 1972 and its phase relations in the B$_2$O$_3$-Fe$_2$O$_3$ system were described\cite{12}.

The magnetic domain properties of FeBO$_3$ were discussed by Scott\cite{4} in 1973 and applications of the FeBO$_3$’s magneto-optical properties were thoroughly discussed by Wolfe in 1970 \cite{1, 6, 13}. FeBO$_3$ was characterized as a material with a spontaneous ferromagnetic moment at room temperature and good transparency in the visible region.
of the spectrum. Ferric borate was proven to be a canted antiferromagnet having a Curie temperature of 348 K and a saturation magnetization of 9.3 G at room temperature\[^{[4]}\].

High quality FeBO\(_3\) single crystal is favored for our ultrafast switching applications. Therefore, the flux growth technique with B\(_2\)O\(_3\)-Fe\(_2\)O\(_3\)-PbO/PbF\(_2\) was used in our lab because of its potential to grow perfectly single crystals.

FeBO\(_3\) is not cubic and has large birefringence for light propagating along the magnetization direction. The simultaneous presence of Faraday rotation and birefringence reduce the maximum obtainable rotation\[^{[14-16]}\]. To compensate the birefringence, two methods could be used. The first one is to stack layers of crystals with alternating birefringent axes\[^{[1,13]}\]. If the thicknesses of successive multilayers are properly chosen, each layer has a fixed rotation degree, where the even-numbered layers are oriented about the c-axis by 90\(^\circ\) with respect to the odd-numbered layers. Therefore, the total structure of n layers will have \(n\chi\) rotation. However, the c-axis of FeBO\(_3\) platelets is out-of-plane, to make one single layer, the platelets need be cut into slices, polished, then glued perpendicular to neighbor layers. To achieve a 45\(^\circ\) rotation, around 100 layers of crystal with around 4\(\mu\)m thick each are needed, which is extremely hard to do and has a high cost. Of course, any multiply of \(\lambda\) could be added to each of these thickness, at the expense of extra absorption without extra rotation.

The second method is to work with randomly oriented nanoparticles\[^{[1,13]}\]. The beam propagating though randomly distributed particles could average the birefringence but keep the Faraday effect. In addition, due to a quantization confinement effect\[^{[17]}\], the optical and other properties of semiconductor Q particles strongly depend on the particle
size and can be dramatically different from the related properties of the bulk material. Characteristic examples of size quantization effects are the changes in absorption spectra of semiconductor particles with changing size: A decrease in size results in a “blue shift” in the absorption spectrum. Therefore, it is expected that the nanoparticle technique could also further improve the transmittance in the visible range.

In our practice, a high energy ball-milling technique was used to create nanoparticles that overcome the birefringence. The FeBO$_3$ platelets were ground into small particles and milled into nanoparticles with a high energy milling machine, and then the nanoparticles were hot-pressed into a new crystal. The new crystal developed this way is expected to have both desired rotation and enhanced transmittance.

### 4.2 FeBO$_3$ Crystals

FeBO$_3$ is a critical phase in our study because of its both advantages of high transmittance and low saturation field, and disadvantage of birefringence. Thus, special effort was put into the synthesizing process of FeBO$_3$. A flux sintering technique was used to grow highly perfect iron borate platelet-like single crystals. The single crystals were tested with X-Ray diffraction and showed the green platelets we obtained have (001) crystallization orientation. Our switching experiments showed that the speed of the switch using FeBO$_3$ platelets has reached 1.2ns with a $\Phi$1 mm circular aperture at the wavelength of 532nm.
4.2.1 Growth of FeBO$_3$ Crystals

The perfect single crystal FeBO$_3$ platelets could be grown by a B$_2$O$_3$-Fe$_2$O$_3$-PbO/PbF$_2$ flux technique $^{[3]}$. Since building a new resistance-heated furnace system is very expensive and time-consuming, a home-made double-layer crucible system was used in our lab. The high purity powders (>99.99%) of iron oxide, boron oxide, lead oxide, and lead fluoride were purchased from Sigma-Aldrich Corp and Alfa-Aesar Corp. The Pt crucible size used was about 1/10 of a standard growth apparatus. The compositions of different components were adjusted according to our new environment. The flux powders were prepared and mixed with the components compositions as in Table 4-1.

<table>
<thead>
<tr>
<th>Components</th>
<th>Fe$_2$O$_3$</th>
<th>B$_2$O$_3$</th>
<th>PbO</th>
<th>PbF$_2$</th>
<th>total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Formula Weight:</td>
<td>159.69</td>
<td>69.62</td>
<td>223.20</td>
<td>245.2</td>
<td>697.71</td>
</tr>
<tr>
<td>Molecule %</td>
<td>4.47</td>
<td>76.15</td>
<td>13.59</td>
<td>5.80</td>
<td>100</td>
</tr>
<tr>
<td>Weight % (g)</td>
<td>0.61</td>
<td>4.56</td>
<td>2.61</td>
<td>1.22</td>
<td>9.00</td>
</tr>
</tbody>
</table>

A 10mL platinum crucible was used to grow the single crystal. To keep the heat uniform during sintering, a second larger ceramic crucible was used to cover the platinum one. Our sintering system is illustrated as in the Figure 4-2.
Before sintering, the powders as in Table 4-1 were mixed and put into a platinum crucible and covered with a top. Then, platinum net was inserted between the crucible and its top. A bigger ceramic crucible was used to hold the platinum crucible tightly with ceramic fiber. Then the ceramic crucible and its top were sealed with cement. Finally, the crucibles were put inside a furnace and sintered according to the temperature illustrated in Figure 4-3. At the end of the growth period, the crucible was turned over and the temperature was decreased at a rate of 0.333 K/min to room temperature.
The platelets grew into larger pieces in the platinum crucible while cooling down to room temperature. Then the platinum crucible and net were washed and cleaned from the rest of flux in hot 5M nitric acid, a lot of green and a little of yellow FeBO₃ crystals were obtained. Figure 4-4 shows a large piece of these crystals whose size is about 7mm long on a wrinkled white paper.
The typical diameter of FeBO₃ crystals is between 2mm to 5mm. One of the largest as in Figure 4-4 has a diameter of 7mm. The thickness is in the range of 10 to 200 µm. Approximately 80% of the growth experiments were successful in yielding crystals of the above described qualities.

**4.2.2 X–Ray Diffraction**

Figure 4-5 shows the X-Ray diffraction of a FeBO₃ platelet single crystal draw by Jade software from Materials Data Incorporated (MDI). The XRD 2θ peak of FeBO₃ at ~37.2° matches (006) characteristic peak of Iron Borate <04-001-8357> in the ICDD database library. There were no other peaks for 2θ from 10° to 70° as shown in Figure 4-5 (a). Figure 4-5 (b) is an enlarged view near the (006) peak.

According to Kotrbova [3], the flux grown crystal has a hexagonal platelets parallel to the (111) plane which is a magnetic easy plane with very low in-plane anisotropy.
Scott\textsuperscript{[4]} confirmed the flux growth has a platelet habit with (111) plane. However, the FeBO\textsubscript{3} crystal we grew shows a hard axis [001] with an easy plane (001) with three-fold symmetry within the plane as in Figure 4-5. The change of growth condition is possibly the main reason causing the orientation change of the single crystal.
Figure 4-5: XRD of a typical FeBO$_3$ crystal (a) 2θ scan from 10-70° (b) enlarged view around the (006) crystal peak
4.2.3 Intrinsic Faraday Effect and Birefringence

The simultaneous presence of Faraday rotation and birefringence in FeBO$_3$ reduce its maximum obtainable rotation. Suppose the magnetization of a sample is $\vec{M}$, the incident light propagates parallel to the unit vector $\vec{k}$, the Faraday rotation angle $F$ is shown as in Equation 4.1 \cite{4} where $F_0$ is the intrinsic Faraday rotation.

$$F = F_0 \frac{\vec{M} \cdot \vec{k}}{|\vec{M}|}$$ \hspace{1cm} \text{Eq. 4.1}

The full rotation is only obtained for propagations in the plane of the platelets. There are two factors deciding the Faraday rotation. One is the platelet thicknesses that is typically less than 100 μm, and the other is the simultaneous presence of Faraday rotation and birefringence, which tend to reduce the maximum obtainable rotation.

For a uniaxial material, the phase retardation $B(\theta)$ at an angle $\theta$ to the optic axis and at a wavelength $\lambda$, can be described by Equation 4.2, where $n_e$ and $n_o$ are the principal refractive indices.

$$B(\theta) = \frac{2\pi}{\lambda} (n_e - n_o) \sin^2 \theta$$ \hspace{1cm} \text{Eq. 4.2}

It is clear from Equation 4.1 and Equation 4.2, that for a small $\theta$, the Faraday rotation varies with $\theta$ and the phase retardation with $\theta^2$, or to say, the Faraday rotation is dominant for a small $\theta$. For the case where $B(\pi/2) >> F_0$, the variation of rotation with the angle $\theta$ can be expressed as in Equation 4.3\cite{13}, where $d$ is the sample thickness and the angles are measured in radians.
The Faraday rotation is the function of $\theta$ and $d$, and it has a damped oscillatory form. Therefore, to obtain the maximum Faraday rotation, one needs to arrange the angle of incidence to coincide with the first maximum in the rotation. Figure 4-6 illustrates the intrinsic Faraday rotation and birefringence as function of $\theta$ in the FeBO$_3$ (001) crystal where $\theta$ is the incident angle related to the optic axis. It is clear than the birefringence increases more than Faraday rotation.

\[
F(\theta) = \frac{F_0 \sin \theta}{B(\theta)} \sin \left( \frac{B(\theta)d}{\cos \theta} \right)
\]  
Eq. 4.3

Figure 4-6: Faraday Rotation and Birefringence of FeBO3 as a function of $\theta$

Figure 4-7 shows the calculated Faraday rotation as a function of $\theta$ at wavelength of 532nm where different color represents different crystal thickness in $\mu$m. It is observed that the first maximum of rotation shifts to the left for thicker crystals. To take advantage of this in switch testing, the angle of incidence should coincide with the first maximum.
Figure 4-7: Calculated Faraday rotation as a function of $\theta$ at 532nm; different color represents different crystal thickness in $\mu$m

Figure 4-8 is the calculated Faraday rotation as a function of thickness in $\mu$m at wavelength 532nm, where different colors represent different incident angles in degrees. It was expected that a maximum rotation about 2.5° at an incident angle around 10° for an 80 $\mu$m thick FeBO$_3$ crystal.
Because the birefringence retardation changes the direction of the right hand of polarized light to the left one periodically, the birefringence of FeBO$_3$ can not be easily compensated by simply putting a compensator behind the FeBO$_3$ sample. Therefore the Faraday rotation will not accumulate in the sample but change with a period of $2\pi$ birefringence phase retardation. Faraday rotation is not proportional to the thickness of the film due to birefringence, and the total Faraday rotation angle is small even for a thick crystal. Therefore it is impossible to reach a $45^\circ$ rotation for a perfect single FeBO$_3$ crystal.

Figure 4-8: Calculated Faraday rotation as a function of thickness at 532nm; different color represents different angle $\theta$
4.2.4 Faraday Rotation and Transmittance

When a light propagates normal to the (001) plane of a FeBO$_3$ platelet, the Faraday effect will not be observed, because the domain magnetizations lie in the plane of the platelet. To observe the Faraday rotation, the platelet needs to be tilted from the light propagation direction as shown in the inset of Figure 4.6, where the angle of incidence coincides with the first maximum Faraday rotation angle for a given thick crystal.

Figure 4-9 shows the testing result of the Faraday rotation on one of our FeBO$_3$ crystals. The saturation magnetic field measured is 20 Oe, and the rotation angle is about $1.5^\circ$ for a 60 $\mu$m thick film at 532nm wavelength.

![Graph showing Faraday Rotation vs Magnetic Field](image)

Figure 4-9: Experimental result of Faraday Rotation angle of a 60 $\mu$m FeBO$_3$ @532nm

The FeBO$_3$ crystals we obtained are transparent and green with a region of low absorption in the visible spectrum.
The transmittance of FeBO$_3$ crystals was tested with a Varian/Cary 100 UV-Vis Spectrometer in at the Penn State Material Research Lab. The data was post-processed with Excel. Figure 4-10 shows the transmittance of the FeBO$_3$. It shows a transmittance peak at around our testing wavelength (532nm YAG Laser). It reached as high as 71% for a 60µm thick film, but fell down to 48% at the wavelength of 611nm. It is obvious that the bandwidth of FeBO$_3$’s transmittance spectrum is a little better than that of BIGG film. The left edge of FeBO$_3$ is at about 470nm while that of a BIGG film is at about 480nm.

Figure 4-10: Transmittance of a FeBO$_3$ crystal

The measured figure of merit (2θ/α) of this 60µm FeBO$_3$ crystal at the wavelength of 532nm is equal to 6.9°
4.2.5 Speed Response of FeBO₃

The switching speed of FeBO₃ is tested at the wavelength of 532nm. Due to its very low saturation field around 20 Oe, FeBO₃ has a potential to reach higher switching speeds. Figure 4-11 show the switching response time of FeBO₃ measured on the oscilloscope is 1.21 ns. As expected, the speed of FeBO₃ crystal is faster than that of the BIGG sample we tested, which was 2.4ns.

Figure 4-11: Switching response time of FeBO₃
4.3 FeBO$_3$ Nanoparticles

As discussed above, the birefringence retardation of FeBO$_3$ changes the direction of the right hand of polarized light to the left one periodically. Therefore, the Faraday rotation will not accumulate in the sample but change with a period of $2\pi$ birefringence phase retardation. In order to eliminate the birefringence and further improve the transmittance, a high energy ball-milling technique was used to synthesize FeBO$_3$ nanoparticles. The randomly oriented nanoparticles are expected to average the birefringence$^{[1, 13]}$. Our simulation shows the nanoparticles do eliminate the birefringence, and concurrently keep the Faraday rotation as shown in section 4.3.1. After milling and centrifuging, the FeBO$_3$ nanoparticle sizes reached down to 30nm so far. The nanoparticles were then hot-pressed and re-formed into an orientation-randomized new crystal. The fabrication technique of FeBO$_3$ nanoparticles were discussed in section 4.3.2.

4.3.1 Simulation of Faraday Rotation of FeBO$_3$ Nanoparticle Film with Different Average Size

Suppose an anisotropic crystal’s a, b, c axes are parallel to x, y, and z axes, respectively, a net magnetization is along ±z, and there is no absorption. Again, the magneto-optical property can be derived by electromagnetic theory if the relative permittivity tensor and thickness of the magnetic material are given. The permittivity tensor is shown in Equation 4.4 where the tensor elements are related to the refractive index by $\varepsilon_1 = n_1^2$, $\varepsilon_2 = n_2^2$, and $\varepsilon_3 = n_3^2$. 
The elliptically polarized normal modes propagate along the z-axis of the crystal as shown in Equation 4.5, where \( k_z = \frac{\omega}{c} n_z \), \( n_z^2 = \frac{1}{2} \left( (\varepsilon_x + \varepsilon_y) \pm \sqrt{(\varepsilon_x - \varepsilon_y)^2 + 4\gamma^2} \right) \), and
\[
\alpha = \frac{2\gamma}{\left| \varepsilon_x - \varepsilon_y - \sqrt{(\varepsilon_x - \varepsilon_y)^2 + 4\gamma^2} \right|}.
\]

By solving the boundary condition with \( \begin{pmatrix} E_x \\ E_y \end{pmatrix} \big|_{z=0} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \), one can find the output of the E components at \( z=l \) as Equation 4.6(14), where \( \kappa = \delta z \), \( \delta = k_z - k_\perp \),
\[
\cos \tau = \frac{(1 - \alpha^2)}{(1 + \alpha^2)}, \quad \sin \tau = \frac{2\alpha}{(1 + \alpha^2)}.
\]

Since the nanoparticles are randomly distributed, it is assumed that light is obliquely incident on an anisotropic crystal, where incident wave vector \( \vec{k} \parallel \vec{z} \) axis. The geometry of the film is depicted as in Figure 4-12, where the angle between the z-axis and c-axis (optical axis of the uniaxial crystal) is \( \theta \), and the angle between the x-axis and the plane containing the z-axis and c-axis is \( \phi \).
In this configuration, there are two polarization directions, the o- (ordinary) and e- (extraordinary) directions. Suppose the ordinary and extraordinary indexes of the crystal are $n_o$ and $n_e$, respectively. The refractive indexes of ordinary and extraordinary waves associated with this wave vector are

\[
\begin{align*}
    n'_o &= n_o, \\
    n'_e &= \left( \frac{1}{n_o^2 \cos^2 \theta} + \frac{1}{n_e^2 \sin^2 \theta} \right)^{-1/2}.
\end{align*}
\]

Therefore, the dielectric tensors are

\[
\begin{align*}
    \varepsilon_e &= n_e^2, \\
    \varepsilon_o &= \left( \frac{1}{n_o^2 \cos^2 \theta} + \frac{1}{n_e^2 \sin^2 \theta} \right)^{-1}.
\end{align*}
\]
Hence, $n_s, n_e$, and $\alpha$ of the principal modes can be obtained easily. We perform a coordinate transform to express incident light in the principal coordinate (whose x- and y-axis are along the o- and e- polarization directions):

$$\begin{pmatrix} E_o \\ E_e \end{pmatrix} = \begin{pmatrix} \sin \phi & -\cos \phi \\ \cos \phi & \sin \phi \end{pmatrix} \begin{pmatrix} E_x \\ E_y \end{pmatrix}$$

After the light goes through a randomly distributed FeBO$_3$ particles determined by Equation 4.6, it is transformed back to original space coordinate:

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix} = \begin{pmatrix} \sin \phi & \cos \phi \\ -\cos \phi & \sin \phi \end{pmatrix} \begin{pmatrix} E_o \\ E_e \end{pmatrix}$$

From the initial vector $\begin{pmatrix} E_x \\ E_y \end{pmatrix}$ and output vector $\begin{pmatrix} E_x' \\ E_y' \end{pmatrix}$, we can deduce the rotation angle and change in polarization state, which includes the orientation of the principle axis of the ellipse and the ratio between the short and long axes of the ellipse.

The M-O effect in a single layer crystal can be easily calculated using the above formalism. The rotation angles refer to the long axes of the ellipses before and after applying the magnetic field. The refractive indices of FeBO$_3$, $n_o$ equals 2.21, and $n_e$, 2.16. The tensors are independent of the applied magnetic field where a saturated magnetic field was assumed. For FeBO$_3$, a saturation field of about 22 Oe is easy to meet.

With the intrinsic Faraday rotation and birefringence formulas mentioned in the section 4.2.3, we analyzed numerically the rotations for FeBO$_3$ particles with different average sizes. In the simulation, we assumed that particles have different average sizes but with the same total length of 100$\mu$m. Figure 4-13 shows the simulated Faraday
rotation results as a function of different average particle sizes, where the input light had a random input angle relative to the particle c-axis direction.

Therefore, for the total length of 100μm FeBO₃ nanoparticle, the Faraday rotation angle we reached in the simulation could be about 16°.

From Figure 4-13(a), we can see that when the average sizes of the nanoparticles are about 100 nm, the birefringence still exists and the total Faraday rotation angle is randomized and at some cases its phase is still reversed. However, when the average sizes become ~ 50nm, Figure 4-13(b) shows that the birefringence is partially eliminated but
the rotation angle is not very stable. When the average sizes become ~ 20nm, the Faraday rotation is stable as shown in Figure 4-13(c). When the average sizes become ~ 10nm, the Faraday rotation is very stable as shown in Figure 4-13(d), which is desirable in our experiment.

### 4.3.2 Fabricate FeBO₃ Nanoparticles via High Energy Ball Milling

In this part, we describe the fabrication of FeBO₃ nanoparticles via a high energy ball milling technique. In order to eliminate the birefringence and further improve the transmittance, FeBO₃ platelets were first ground and ball-milled into nanoparticles. A high energy Spex 8000 ball milling machine was purchased and used for this purpose, as shown in Figure 4-14.

![Spex 8000M Mixer/Mill machine](image)

Figure 4-14: Spex 8000M Mixer/Mill machine
In our experiments, 1 g FeBO₃ powder with an average size around several microns was loaded into the steel vial. The milling time was around 90 hrs. The size of FeBO₃ particles were substantially reduced from the micron size to the smallest size around 20 nm range, as shown in Figure 4-15.

To our knowledge, this is the smallest size FeBO₃ particles ever reported. Generally, small nanoparticles have a tendency to form larger particles due to the agglomeration effect. The small particles were then centrifuged and separated successfully from large particles. After milling and centrifuging, the FeBO₃ nanoparticles’ sizes reached down to 30nm so far. The nanoparticles are to be hot-pressed and re-formed into an orientation-randomized new crystal. It is expected that the nanoparticles approach will overcome the birefringence, improve the transmittance, and concurrently keep the same switching speed. Further tests need to be conducted on these FeBO₃ nanoparticles.
When the nano-particle size is small enough, the quantum confinement effect will generate a blue shift \cite{17} in the absorption spectrum. We believe that similar blue shift effect due to the quantum confinement should exist in ferrite borate materials because FeBO$_3$ have a direct bandgap, which is similar to the bandgap of the semiconductor materials like silicon. The blue shift of FeBO$_3$ should improve the transmittance too.

\textbf{4.4 Conclusion}

Iron Borate (FeBO$_3$) is far superior in terms of figure of merit (Faraday rotation to absorption ratio) in the visible spectrum at room temperature to other materials. A flux sintering technique was used in our lab to grow Iron Borate. Our test shows the green platelet we grew had good (001) crystallization and the switching test reach a speed of 1.2ns. However, the iron borate crystal structure is not cubic and has large natural

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{SEM_image.png}
\caption{SEM image of FeBO$_3$ nanoparticles}
\end{figure}
birefringence, which impedes the light propagated along the magnetization. Our simulation shows the nanoparticles could eliminate the birefringence, and concurrently keep the Faraday rotation. A high energy ball-milling technique was used and FeBO$_3$ nanoparticles were synthesized into about 30nm size. The nanoparticles were then hot-pressed to form a new random-oriented crystal. Further experiments and investigations are needed and we expect new crystal could overcome the birefringence, improve the transmittance, and still have the same switching speed.

### 4.5 References


Chapter 5

Synthesis of New BYIGG Nanoparticle and their Optical and Magneto-optical Properties

5.1 Introduction

Bismuth substituted Yttrium Iron garnets such as Bi$_{1.8}$Y$_{1.2}$Fe$_5$O$_{12}$ (BYIG) are attractive magneto-optical materials that possessing enhanced Faraday Effect. The Faraday rotation angle can be as high as 2$^\circ$/μm/per Bi content at wavelength of 633nm [1, 2]. The garnet film has been prepared with deposition processes, which require expensive substrates, e.g. GGG (Gadolinium Gallium Garnet). BYIG nanoparticles have been studied and synthesized by coprecipitation and annealing processes in the recent years [3-11]. The nanoparticles are mixed with solvents and milled, and then coated on a substrate. The advantages of a nanoparticle film over sputtering are its low cost and high yield. Currently, the Faraday rotation of BYIG nanoparticles have been reported over a range from 8$^\circ$/cm to 10$^3$°/cm depending on the size and the weight density [4, 6, 8, 9], and the figure of merit (2θ/α) reached over a range from 0.2$^\circ$ to 9.6$^\circ$ depending mainly on the particle size [3, 11]. In contrast, bulk BYIG crystal has much higher rotation as high as 2$^\circ$/μm at wavelength of 532nm but with a very low transmittance. Because the BYIG nanoparticles were mixed with transparent solvents, the films made of nanoparticles sacrifice the Faraday rotation per thickness and gain transmittance.
In this chapter, the synthesis of new garnet BYIGG nanoparticles (Bi$_{1.8}$Y$_{1.2}$Fe$_4$Ga$_1$O$_{12}$) by coprecipitation and annealing processes is investigated. Similar to our BIGG films case, Gallium was doped and substituted a partial of iron ions. The new made powders possess more yellowish color than BYIG powders. The particle was milled and coated on a glass and film’s optical and magneto-optical properties were checked. Theoretical investigation on scattering was also addressed and, it is explained how the scattering was related with the nanoparticle sizes.

5.2 Synthesis of New BYIGG Nanoparticles

5.2.1 Preparation of BYIGG Nanoparticles

Figure 5-1 shows the flowchart of preparation of the new BYIGG (Bi$_{1.8}$Y$_{1.2}$Fe$_4$Ga$_1$O$_{12}$) nanoparticles. First, the Aqueous solutions of Bi(NO$_3$)$_3$, Y(NO$_3$)$_3$, Ga(NO$_3$)$_3$, and Fe(NO$_3$)$_3$ were mixed where the ratio of cations was kept to the same composition as that of Bi$_{1.8}$Y$_{1.2}$Fe$_4$Ga$_1$O$_{12}$. The solution was then mixed with NH$_4$OH solution and stirred at room temperature. The concentration was adjusted so that the pH of the mixed solution reached 10.5 at the end of the coprecipitation. The obtained slurry was then filtered with VWR 413 filter paper and dried at 100°C for about 10 hour. At last, the dried material was annealed at 650°C for 4 hours.
After annealing, the material became powder as shown in Figure 5-2. It is clearly shown that the BYIGG nanoparticles we made were more yellowish than the BYIG nanoparticles. Based on the results in the chapter 3, we know the transmittance of BIGG film is much better than BIG film. Therefore, the gallium doping could improve the transmittance of iron garnet materials.
5.2.2 Ball-milling and Span-coating Processes

The BYIGG nanoparticles have been mixed with different solvents for ball-milling. Three different solvents were tested and evaluated. The solvent of methyl-methacrylate (MMA) with poly-MMA was easy to be solidified for long time milling while the Vertrel XF solvent was too corrosive and our powders were contaminated badly. A cyclohexanone solvent works well currently. When the nanoparticles were mixed the cyclohexanone solvent, the contamination was low.

The high energy ball-mill machine and ball vial sets used were from Spex as shown in Figure 4.13. A Specialty Coating System P-6000 span-coater was used to coat the milled-mixtures on Corning 1737 glass substrate. The coated film was then dried at 80°C for 1 hour in an oven.

5.3 Properties of BYIGG Nanoparticles

5.3.1 X-Ray Diffraction

Figure 5-3 shows the X-Ray Diffraction (XRD) of BYIGG nanoparticles, where the characteristic peaks of Bi\(_{1.5}\)Y\(_{1.5}\)Fe\(_5\)O\(_{12}\) are for comparison. Even though the characteristic peak of our new BYIGG nanoparticles are very close to that of BYIG
nanoparticles, we can still clearly see an extra peak at such as about 22.5° and 28°, which were distinguished from BYIG material.

![XRD of BYIGG nanoparticles](nano_#005_BYIGG_Nov16_2006.raw)

Figure 5-3: XRD of BYIGG nanoparticles

### 5.3.2 Nanoparticle Size Measurement

The Zetasizer Nano series nano-sizer at the Penn State Material Research Lab was used to measure the particle sizes during the ball-milling process. Figure 5-4 shows a typical particle size distribution after 10 hours milling.
5.3.3 Faraday Rotation and Figure of Merit

Faraday rotation was measured at wavelength 532 nm in our Lab. The nanoparticles were milled for 40 hours and the estimated size is about 30 nm. The Faraday rotation angle was about 2.5° for a 500 μm thick film, while the transmittance was about 1% at the wavelength of 532 nm. The Figure of merit was found to be 2.1° which was among the reported range from 0.2° to 9.6° [3, 11]. The reason for the low Figure of merit is possibly the contamination which we need more investigation to clarify.
5.4 Theoretical Explanation

Generally, optical scattering exists in small particle media. Under the condition that the size of BYIGG nanoparticles is much less than the wavelength of the incident light (532nm), Rayleigh scattering model is a good approximation of theoretical estimation of the optical scattering loss [12-14].

According to the classic scattering theory of electromagnetic waves, the scattering cross-section of each particle can be described as in Equation 5.1, where \( V \) is the volume of each particle in \( \text{m}^3 \), \( n_M \) and \( n_D \) are the refractive indices of the magnetic nanoparticles and the surrounding dielectric material, respectively.

\[
\sigma_s = \frac{24\pi^3 V^2}{\lambda^3} \left| \frac{n_M^2 - n_D^2}{n_M^2 + 2n_D^2} \right| \quad \text{Eq. 5.1}
\]

Assume that the volume density of the particles is \( D_v \), for a unit length of the waveguide, the total number of particles is \( D_v/V \). Therefore, the loss parameter in \( \text{m}^{-1} \) due to scattering is as presented in Equation 5.2:

\[
\alpha_s = \frac{\sigma_s D_v}{V} = \frac{24\pi^3 D_v}{\lambda^4} \left| \frac{n_M^2 - n_D^2}{n_M^2 + 2n_D^2} \right| \quad \text{Eq. 5.2}
\]

Under saturation magnetic field, a typical value of Faraday rotation angle of BYIG bulk film can be 2.5°/\( \mu \text{m} \) at 532nm from our estimation in chapter 2. Therefore, in order to generate a 45° Faraday rotation, the length of a BYIG-imbedded optical waveguide has to be \( d = 45/(\Phi \Phi D_v) \). As an example, for a volume density of the particles \( D_v = 5\% \), the required waveguide length is approximately 36\( \mu \text{m} \). The scattering loss can be derived from Equation 5.3 where the volume density is eliminated because the increase
of particle density will increase scattering loss but on the other hand a reduced the thickness needed.

\[ A_s = \exp\{-\alpha_s d\} = \exp\left\{ -\frac{24\pi^3 D_v}{\lambda^4} \left( \frac{n_{D}^2 - n_{M}^2}{n_{M}^2 + 2n_{D}^2} \right) \frac{45}{\Phi_D D_v} \right\} \]

Eq. 5.3

\[ = \exp\left\{ -\frac{24\pi^3}{\lambda^4} \left( \frac{n_{M}^2 - n_{D}^2}{n_{M}^2 + 2n_{D}^2} \right) \Phi_F \right\} \]

If the refractive indices of the solvent \( n_0 \) range from 1.5 to 2.0, in order to generate a 45° Faraday rotation, the scattering transmission loss (defined as \(-\ln(A_s)\)) of the optical waveguide caused by the BYIG particle with solvent is shown in Figure 5-5.

Figure 5-5: The scattering loss \(-\ln(A_s)\) of the BYIG particles with solvent whose refractive index from 1.5 to 2.0
According to Figure 5-5, in order to achieve 45° Faraday rotation and scattering loss <1 for a \( n_0 = 1.6 \) solvent, the required size of nanoparticles should below 30nm approximately.

On the other hand, if the absorption coefficient of the bulk BYIG is \( \alpha_m \), then the excess optical absorption coefficient \( \alpha_a \) due to the solvent can be expressed as in Equation 5.4.

\[
\alpha_a = \frac{9\alpha_m D_D}{\left(1 + \frac{n_D^2}{n_D^2 + 2}\right)^2}
\]

Eq. 5.4

At the wavelength of 532nm, the absorption coefficient for BYIGG garnet is quite large, a typical optical absorption coefficient for BYIG is approximately \( \alpha_m = 4 \times 10^3 \) [cm\(^{-1}\)]\(^\text{[15]}\). The absorption is as shown in Equation 5.5.

\[
A_a = \exp\left\{-\alpha_a d\right\} = \exp\left\{-\frac{9\alpha_m D_D}{\left(1 + \frac{n_D^2}{n_D^2 + 2}\right)^2} \Phi_F D_D \right\} = \exp\left\{-\frac{9\alpha_m}{\left(1 + \frac{n_D^2}{n_D^2 + 2}\right)^2} \Phi_F \right\}
\]

Eq. 5.5

To generate a 45 degree Faraday rotation, the absorption transmission loss (defined as -\( \ln(A_a) \)) caused by the BYIG particle with solvent is shown in Figure 5-6.
By comparing Figure 5-5 and Figure 5-6, it is clear that the absorption will be dominant when the particles’ size is smaller than 20nm -30nm. The particle sizes affect the scattering but not the absorption. Therefore, the transmittance loss will not significantly decrease after the particle size decreased to 17nm as shown in Figure 5-7.

To further reduce the total transmittance loss, new BYIGG nanoparticles with lower absorption are required. From the color of new particles and the transmittance coefficient of BIGG films in chapter 3, we can expect an order lower absorption loss from new BYIGG nanoparticles. When using a reasonable absorption coefficient $\alpha_m = 4 \times 10^2$ and

![Figure 5-6: The absorption loss $-\ln(A_a)$ caused by the BYIG particle with solvents whose refractive index from 1.5 to 2.0](image-url)
combine the scattering loss, the total transmission loss \(-\ln(A_a)\) caused by the BYIGG particle with solvent can be shown as in Figure 5-7.

Therefore, by combining the effects of scattering and excess absorption, and taking into consideration the efficiency of Faraday rotation, in order to generate a 45° Faraday rotation and the transmittance loss <1 for solvent with a refractive index \(\approx 1.5\), the required average size of BYIGG nanoparticle should be roughly about 22nm.

Figure 5-7: The total transmission loss \(-\ln(A_a)\) caused by the BYIGG particle with solvent.
5.5 Conclusion

The advantages of magneto-optical nanoparticle film over sputtering are its low cost and high yield. A new nanoparticle garnet material BYIGG was synthesized, milled, and spin-coated on a Corning 1737 glass to improve the transmittance and figure of merit of nanoparticle film. Theoretical explanation of scattering was investigated and how the scattering is related to the nanoparticle size was explained. In order to generate a 45 degree Faraday rotation and the transmittance loss <1 for the refractive index equals 1.5 solvent, the required average size of BYIGG nanoparticle should be roughly about 22nm. The size of the nanoparticles, X-ray diffraction, transparency, and Faraday rotation angle were measured. The Faraday rotation angle was of about 2.5 ° for a 500 μm thickness film, and the transmittance at 532nm was about 1 %. The figure of merit (2θ/α) was found to be 2.2°, which is among the reported ones from 0.2° to 9.6°. The possible reason for the low figure of merit is contamination, which needs more experiment and investigation to verify.

5.6 References


6.1 Introduction

If the driving circuit response time is larger than the films/crystals’ response times, the switching speed will be limited by the driving circuit board. In this part, we focus on the design of the driving circuit board, including the design of the miniature driving coil, and RF circuit board to insure that the circuit response is faster than the testing result.

In modulator and isolator devices, the magnetic field could be generated by a transmission line or a microstrip [1-5] where the microscope objectives were used to focused the beam into a waveguide magneto-optical crystal. Because the magnetic field under the microstrip drops off inversely as the square of the distance [6, 7], the output aperture size is usually limited to 10 to 25μm. For our free space ultrafast application project, the aperture size for a free space beam is required to be larger than 300μm and the magnetic field should be as uniform as possible. Therefore, a transmission line or microstrip is not feasible any more. In this study, a miniature coil was used. The coil should be made in such a way that it is of the minimum possible size, it generates enough magnetic field, and still allows all light to pass through the center of the coil.

On the other hand, a coil has self–inductance and resistance, and the other circuit parts also have residual resistances and residual inductances. The total circuit will have a
time constant $\tau = L/R$ depending on the total inductance $L$ and resistance $R$, which may limit the final speed of the switch even if the material is fast enough. Therefore, the total time constant $\tau$ needs to be designed smaller than the film’s response time. This means, the inductance $L$ should be small or the resistance $R$ should be big. But the resistance is limited by current supply capacity, which leaves us the only choice to reduce the total inductance as small as possible. This means, the coil should have less turns.

Based on the above discussion, it is a challenge to design a coil with both desired magnetic field and a small inductance. If not properly designed, the speed will be limited by the circuit parameters. Trade-offs between the magnetic field and the response time was made in our design. Our design of magnetic fields of coils are discussed in detail in section 6.2, the coil response parameter is investigated and discussed in section 6.3, and the RF driving circuit design is included in 6.4.

### 6.2 Magnetic Fields Generated by Coils

Both a solenoid coil and a spiral coil were used to generate the magnetic field in our switch experiments. To generate a higher magnetic field with a fixed current, the coil was designed to have several (3 to 10) turns. A solenoid coil was wound by hand and it had 5-turns and was 1mm in diameter. Specifically, an AWG #42 wire was first wound on a <1mm drill, and then the drill was removed. A spiral coil was designed by AutoCAD and fabricated with lithography technique where Au was deposited on a film/crystal.
6.2.1 Solenoid coil

The magnetic field of a solenoid coil can be calculated directly from Equation 6.1 where \( N \) is the total turns of wires, \( I \) the current, and \( L \) the length of coil.

\[
H = \frac{N I}{L} \quad \text{Eq. 6.1}
\]

Table 6-1 show one of specifications of a solenoid coil we made.

<table>
<thead>
<tr>
<th>Turn</th>
<th>Diameter of wire loops</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calculated inductance</td>
<td>0.82 mm</td>
</tr>
<tr>
<td>Calculated magnetic field</td>
<td>0.07 mm (AWG 42)</td>
</tr>
<tr>
<td>Calculated inductance</td>
<td>50 nH</td>
</tr>
<tr>
<td>Calculated magnetic field</td>
<td>50 Oe/A</td>
</tr>
</tbody>
</table>

6.2.2 Spiral coil

For an arbitrary coil, the magnetic field was simulated based on the electromagnetic field theory\([6, 7]\). The magnetic field on a point created by a current wire is the integration of all fields contributed by infinitesimal piece of the current wire as described in Equation 6.2.

\[
\vec{H} = \int_{L} \frac{Id\vec{l} \times \vec{i}_{QP}}{4\pi r_{QP}^2} \quad \text{Eq. 6.2}
\]

Not losing generality, suppose an arbitrary shape current wire \( L \) is located in X-Y plane. Let \( I \) be the current used in the artistry wire \( L \), \( Q \) a point on the wire, \( d\vec{l} \) an infinite length along the wire \( L \), \( \vec{i}_{QP} \) the unit vector from the points \( Q \) to \( P \), \( r_{QP} \) the
distance between the points Q to P, and $\vec{H}$ the magnetic field on a point P(x,y,z).

Figure 6-1 illustrates the schematics of the magnetic field generated by infinitesimal current piece $rd\phi$.

Figure 6-1: Schematics of the magnetic field generated by infinitesimal current piece $rd\phi$. (a) 3D view; (b) the wire position on x-y plane

Figure 6-2 shows the magnetic field generated by 10 turn circular wires. The 10 circular wires were not shown on the figure, but have the center on the figure center. The inner circle’s diameter is 300μm, the line width 10μm, the line-gap is 5μm, and the outer circle 435μm. With 1A driving current, the center area can reach 400 Oe as shown in Figure 6-2.
During the deposition of the coil on a film, a thin SiO\textsubscript{2} layer with a thickness of 50 nm was deposited as a nonmagnetic isolation layer and then a gold layer was deposed on the top of it. Figure 6-3 shows the mask pattern designed with AutoCAD and VB script.

Figure 6-2: Magnetic field generated by 10 turn circular wires with 1A current
A designed spiral or circular mask was used for lithography to pattern the required coil structure. A miniature gold coil was sputtered and etched on the top of garnet film. Figure 6-4 shows a single Au wire deposited on a garnet film.

Figure 6-3: Spiral mask pattern designed with AutoCAD and VB script

Figure 6-4: Au coil deposited on a garnet film
6.3 Response Time of a Spiral Coil

In the switching response test, the total inductance needs to be calculated where the time constant will not limit the switching response. A 10-turn circular wire was used to simulate the inductance, resistance, and the time constant.

6.3.1 Inductance

The structure of a thin-coil model was used to calculate the inductance using the model shown in Figure 6-5, where

\[ a = \text{mean radius of the turns} \]
\[ b = \text{axial dimension of the cross section} \]
\[ c = \text{radial dimension of the cross section} \]
\[ N = \text{total number of the turns} \]

\[ L \] is the inductance of a circular coil with rectangular cross section \[^{8,9}].\]
A thin coil’s inductance can be found from Equation 6.3, where “a” in cm and L in $\mu$H, and $K' = K - k$

$$L = 0.019739 \left( \frac{2a}{b} \right) N^2 a_{cm} K'$$ \hspace{1cm} \text{Eq. 6.3}

In our 10-turn circular coil model, the width of the line is 10$\mu$m, the line gap is 5$\mu$m, the deposition thickness of gold layer is 2$\mu$m, and the diameter of the inner circle is 300$\mu$m.

After calculation and looking into the table$^8$, we found,

$$L = 0.02 \mu H = 20nH$$

The inductance of 20nH of a 10-turn coil is a relatively small value and is good for our RF circuit design.

Figure 6-5: A circular coil with a rectangular cross section
6.3.2 Resistance

A same 10-turn spiral wire is used to calculate the resistance of the coil.

A typical thin film metal structure such as gold has a measurable resistance due to the cumulative sensitivities of the metals. This resistance is commonly related to sheet resistance. The unit of measure of sheet resistance is ohms per square (Ω/□). For a circuit consisting of a single metal layer, its total resistance can be calculated by Equation 6.4

\[
R = \frac{\text{trace length}}{\text{trace width}} \times \text{sheet resistance} = \frac{L_{\text{trace}}}{W_{\text{trace}}} \times R_{\text{sheet}}
\]

Eq. 6.4

When a 2µm thick gold layer is sputtered as the conducting wire, the sheet resistance is about 10-11mΩ/□. The circle diameters of the 10-turn coil are from 300µm to 435µm with a step increase 15µm for each outer turn. If the line width is 10µm, the total resistance is approximately

\[
R = \frac{\pi(300 \div 15 \div 435)\mu m}{10\mu m} \times 10.5m\Omega = 1.04 \div 0.06 \div 1.5\Omega
\]

Since R can be serially configured, the total R≈12.5Ω

The resistance can be increased by serially connecting another resister. However, it is limited by current supply capacity. If the total resister is 50 Ω, a pulsed power supply with a 200 volt output can generate 4A current.
6.3.3 Circuit Time Constant

The time constant is shown in Equation 6.5, where \( R \) is the resistance and \( L \) is inductance of the RF circuit.

\[
t = \frac{L}{R}
\]

Eq. 6.5

10 turns of a spiral or 10 turn circles should have an inductance of 20nH. For inductance 20nH and resistance 50 \( \Omega \), the response time can be estimated as:

\[
t = \frac{L}{R} = 0.4 \text{ ns}
\]

This will not limit the film’s switching response time.

6.4 RF Driving Circuit Design

The circuit board is also a key part of designs because the residual conductance and resistance need be considered, the RF driving circuit design and the circuit board response test were done with the assistance of Chia-En Yang.

6.4.1 Driving Circuit Design

The simulation is done with Agilent Advanced Design System (ADS). The inductance of the coil is assumed to be <50 nH. This inductance, though larger than the theoretical value, was used to account for other parasitic inductance on the circuit.

Figure 6-6 shows the electric circuit diagram of the driver circuit board. Increasing \( R \) will reduce the rise time of the current in \( L \), which is very desirable.
However, this will also reduce the amount of DC current in L since it is proportional to the ratio R2/R. Increasing R2 will solve this problem but will cause the DC effective impedance to be different from 50 ohm. R1 and R2 work together to reduce the real-time signal reflection from the circuit.

---

![Electric Circuit Diagram](image)

**Figure 6-6:** The electric circuit diagram of the driver circuit board

Another important design issue is the size of the actual circuit board. To qualify as a lumped circuit, the total length of the PCB should be smaller than 1/6 of the length of the signal rising edge. This is calculated to be about 35 mm. Also, a ground plane is needed to reduce parasitic inductance of the traces, and the traces on the board are designed to have a characteristic impedance of 50 ohm to reduce reflection. The PCB design is shown in Figure 6-6 where the requirements of the SMD resistors are:

- Maximum applied voltage > 120 V;
- Power > 0.2 W;
• Parasitic inductance < 1 nH.

6.4.2 Response Testing of the Driving Circuit

Figure 6-7 is the picture of the finished the driver circuit board.

Figure 6-7: The picture of the designed driver circuit

Figure 6-8 shows electronic switching time of the driver circuit has a rise time (standard definition: 10% to 90%) within less than 1 ns.
6.5 Conclusion

In this chapter, we calculated the required magnetic fields, the inductance, the resistance of coils, and designed a RF circuit board that meet the requirement of circuit time constant. The response time constant of new designed RF circuit is less than 1ns. The circuit we designed was used in the switch test for various magneto-optical materials.

6.6 References


Chapter 7
Theoretical Optimization of Aperiodic Multilayer Structures with a Genetic Algorithm

In this chapter, the optical and magneto-optical properties of one-dimensional non-periodic (aperiodic) multilayer structures were theoretically studied and designed by means of a genetic algorithm. The purpose is to provide a method for the computation of aperiodic multilayer structures that enhances both transmittance and the Faraday Effect, which has not been studied in the past. The 4X4 matrix approach\(^{[1,2]}\) is used to find the transfer function between neighbor layers, a genetic algorithm is used to seek the layer thicknesses of the best aperiodic structures, and a pattern function is used to compare the different structures. Quantitative computer simulation results are provided in the materials composed of bismuth-substituted yttrium–iron–garnet and dielectric (SiO\(_2\)) films.

7.1 Introduction

The optical and magneto-optical properties of one-dimensional photonic crystals (1D_PCs) of periodic multilayer structures with one or more defects has been studied by researchers theoretically\(^{[1,3-10]}\) and experimentally\(^{[4,9]}\) in cases where at least one of the component materials is magnetic. Because of their considerable localization of light, these media exhibit both enhanced Faraday rotation and transmittance at selected wavelengths. In these cases, periodic structures with one or more defects were proposed,
but no studies had been performed for the general case of aperiodic structure due to the quantity of unknown parameters needed to determine such a structure.

In this study, a unique technique for 1D PCs of aperiodic multilayer structures was studied theoretically. These aperiodic structures show potential to possess similar or higher Faraday rotation and transmittance (performance) when optimized.

As we know, given a predefined multilayer system, the optical or magneto-optic properties can be derived with a 4X4 matrix approach \[1-5\] based on Maxwell’s equations. Each layer is characterized with a matrix associated with the complex permittivity tensor of the material and its thickness. The performance of a structure depends on how the structure is designed.

Suppose an arbitrary multilayer structure with layer thicknesses of
\[
[d_{M1}d_{Di1}d_{M2}d_{D2} \ldots d_{Mi}d_{Di} \ldots d_{MN}d_{DN}]
\]
where \(d_{Mi}\) and \(d_{Di}\) are the \(i^{th}\) layers’ thicknesses of magnetic and dielectric materials respectively and \(i=1, 2, \ldots, N\). All layers’ thicknesses are unknown parameters that need to be determined in order to obtain the overall properties of a structure. A randomly picked structure with arbitrary thicknesses of each layer will most likely result in poor performance, except the specially designed periodic structures with one or more defects as mentioned above.

Our simulation began with a 2\(\mu m\) 1D-PC, and the thickness precision of a single layer was set to 1nm. The higher the precision, the longer the calculation time needed. To meet 1nm precision requirement, the total 2\(\mu m\) crystal need be digitalized into 2,000 layers, where each layer is 1nm thick as shown in Figure 7-1. Initially, the layers are randomly assigned as one of two materials: Bi-YIG and SiO\(_2\) materials. With the
optimization technique discussed later, the tiny neighbor layers with the same component material are grouped as a thicker either magnetic or dielectric material layer.

The total number of all possible structures is \(2^{2000} \approx 1.15 \times 10^{602}\) because of the 2000 tiny layers and 2 component materials. It is nearly impossible to calculate and compare all \(1.15 \times 10^{602}\) structures using an exhaustive approach because even if each structure requires 0.001 second of computation time, with 64 parallel high performance computers, it will still take \(5.7 \times 10^{589}\) years. Therefore, some optimization method must be utilized.

In response to this demand, a genetic algorithm is developed and used to optimize a structure without exhaustively calculating all possibilities. A pattern function is applied at the same time to evaluate and compare the transmittance and the Faraday effect of different structures.

Genetic Algorithms (GA) are global numerical-optimization approaches different from gradient and other methods\(^{[11-14]}\). By modeling natural evolution and genetic recombination, the algorithm is suitable for optimization of procedures with a large number of discrete parameters. Because of the nature of the multilayer system in this study, a genetic algorithm is suitable to optimize the corresponding optical and magneto-optical properties. With the algorithm, the best aperiodic structures are found with much fewer calculations because of this algorithm’s unique optimization capability.

In section 7.2 Faraday rotation and transmittance of multilayer structures of magneto-optical garnet materials are discussed. Section 7.3 presents the background of Genetic Algorithms and how to find the desired structure system for best optical and magneto-optical properties. Section 7.4 discusses the simulation results of aperiodic multilayer structures, while conclusions and a summary are given in section 7.5.
7.2 Faraday rotation and transmittance in multilayer structures of magneto-optical garnet materials

Suppose a 2\(\mu\)m film composes 2000 tiny layers and each layer extends boundlessly in the X-Y plane. Each tiny layer has a thickness of 1nm. 2000 tiny layers are stacked in the Z-axis direction in a random sequence so as to form the random multilayer film with total thickness equal to 2.0\(\mu\)m as shown in Figure 7-1. Either Bi:YIG or SiO\(_2\) material is initially assigned to each tiny layer randomly. While the structure is optimizing, the neighboring layers are grouped to a single material gradually. A typical structure can be represented

\[
\left[ \begin{array}{cccccccc}
1 & 1 & 1 & 1 & 0 & 0 & 0 & 0
\end{array} \right]^{2000} \]

where 1(true) and 0 (false) represent the magnetic and dielectric tiny layers, respectively.

\(d_{M1}, d_{D1}, \ldots, d_{Di}, d_{M2}, \ldots, d_{MN}\) is another expression for the thickness distribution of the magnetic and dielectric layer where the subscript M stands for the Bi:YIG layer and D the dielectric SiO\(_2\) layer.
As we know, the transmittance and Faraday rotation from a single layer film depend on the film thickness $d$, the complex permittivity tensors of the material, and the wavelength $\lambda$ of normal incident light. A 4X4 state matrix approach is used based on Maxwellian electromagnetic field theory. A single magnetic layer’s state vector equation has been rewritten for easy calculation in Matlab as shown in Equation 7.1. The state vector is defined as $\boldsymbol{\tau}(Z) = (e_x \quad e_y \quad h_x \quad h_y)$ and $\Phi$ is a 4X4 matrix determined by matrix M and S, where $k_p = (\omega/c)\sqrt{\varepsilon_p}$, $k_n = (\omega/c)\sqrt{\varepsilon_n}$ are the wave numbers, and $\varepsilon_p = \varepsilon_1 + \varepsilon_2$, $\varepsilon_n = \varepsilon_1 - \varepsilon_2$ are the right and left circularly polarized permittivities, and right and left phase $\delta_p = k_p d$ and $\delta_n = k_n d$. A single dielectric (D) layer’s 4X4 matrix can be found when substituting $\varepsilon_D$ into both $\varepsilon_p$ and $\varepsilon_p$. 

Figure 7-1: A multilayer structure with a 1nm single layer thickness
When a multilayer occupies the spatial region of \( Z < z < 0 \), the tangential components of the electromagnetic fields are continuous at all interfaces. Therefore, the state vector is also continuous. For a light entering at \( z = 0 \) and exiting at \( z = Z \), the state vector can be rewritten as Equation 7.2

\[
\tau(Z + d) = \Phi \tau(Z)
\]

where

\[
\Phi = MSM^{-1}
\]

\[
M = \begin{bmatrix}
1 & 1 & 1 & 1 \\
-i & -i & i & i \\
i\sqrt{\varepsilon_p} - i\sqrt{\varepsilon_p} - i\sqrt{\varepsilon_n} + i\sqrt{\varepsilon_n} \\
\sqrt{\varepsilon_p} - \sqrt{\varepsilon_p} - \sqrt{\varepsilon_n} + \sqrt{\varepsilon_n}
\end{bmatrix}
\]

Eq. 7.1

\[
S = \begin{bmatrix}
e^{i\delta_p} & e^{-i\delta_p} & e^{i\delta_n} & e^{-i\delta_n}
\end{bmatrix}
\]

When a multilayer occupies the spatial region of \( Z < z < 0 \), the tangential components of the electromagnetic fields are continuous at all interfaces. Therefore, the state vector is also continuous. For a light entering at \( z = 0 \) and exiting at \( z = Z \), the state vector can be rewritten as Equation 7.2

\[
\tau(z = Z) = \Phi \tau(z = 0)
\]

and

\[
\Phi = \Phi_{M1} \Phi_{D1} \cdots \Phi_{M} \Phi_{D} \cdots \Phi_{MN}
\]

Eq. 7.2

Where the state vectors \( \Phi_{M1} \Phi_{D1} \cdots \Phi_{M} \Phi_{D} \cdots \Phi_{MN} \) corresponds to each layer structure with thickness of each layer as \( d_{M1}, d_{D1}, \ldots, d_{M}, d_{D}, \ldots, d_{MN} \). With Equation 7.1, the state matrix \( \Phi \) can be obtained for each structure. The multiple sequences of the state matrices \( M \) and \( G \) in Equation 7.2 correspond to the sequence of alternating magnetic and dielectric layers.
The state vector in the exterior space of $z<0$ is given by the sum of the ingoing TM light and the reflected TM and TE lights. The state vector in the exterior space of $z>Z$ is given by the sum of the outgoing TM and TE lights only as in Equation 7.3. where $C_1$, $C_2$, $C_3$, and $C_4$ are coupling coefficients which need to be determined representing the field amplitudes of the reflected TE and TM light and the transmitted TM and TE lights respectively. By substituting Equation 7.3 into Equation 7.2, $C_1$, $C_2$, $C_3$, and $C_4$ can be solved for from Equation 7.4.

$$\tau(z \leq 0) = \begin{bmatrix} 1 \\ 0 \\ 0 \\ 1 \end{bmatrix} e^{ikz} + C_1 \begin{bmatrix} 1 \\ 0 \\ -1 \\ 0 \end{bmatrix} e^{-ikZ} + C_2 \begin{bmatrix} 0 \\ -1 \\ -1 \\ 0 \end{bmatrix} e^{-ikZ}$$

Eq. 7.3

$$\tau(z \geq Z) = C_3 \begin{bmatrix} 1 \\ 0 \\ 0 \\ 1 \end{bmatrix} e^{-ik(z-Z)} + C_4 \begin{bmatrix} 0 \\ 1 \\ -1 \\ 0 \end{bmatrix} e^{ik(z-Z)}$$

$$\tau(z \geq Z) = \Phi \tau(z \leq 0)$$

$$\Phi = \begin{bmatrix} \phi_{11} & \phi_{12} & \phi_{13} & \phi_{14} \\ \phi_{21} & \phi_{22} & \phi_{23} & \phi_{24} \\ \phi_{31} & \phi_{32} & \phi_{33} & \phi_{34} \\ \phi_{41} & \phi_{42} & \phi_{43} & \phi_{44} \end{bmatrix}$$

Eq. 7.4

$$\begin{bmatrix} \phi_{11} - \phi_{14} & \phi_{12} + \phi_{13} & -1 & 0 \\ \phi_{21} - \phi_{24} & \phi_{22} + \phi_{23} & 0 & -1 \\ \phi_{31} - \phi_{34} & \phi_{32} + \phi_{33} & 0 & 1 \\ \phi_{41} - \phi_{44} & \phi_{42} + \phi_{43} & -1 & 0 \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \\ C_3 \\ C_4 \end{bmatrix} = \begin{bmatrix} \phi_{11} + \phi_{14} \\ \phi_{21} + \phi_{24} \\ \phi_{31} + \phi_{34} \\ \phi_{41} + \phi_{44} \end{bmatrix}$$
The transmittance $T$ and Faraday rotation $\theta_F$ then can be found from Equation 7.5 when the structure of an aperiodic multilayer film is known where $0 \leq T \leq 100\%$ and $0 \leq \theta_F \leq 90^\circ$

$$T = |C_3|^2 + |C_4|^2$$

$$\theta_F = \tan^{-1}\left(\frac{C_3 C_4^* + C_4 C_3^*}{C_3 C_3^* - C_4 C_4^*}\right)/2$$

$$\eta_F = \tan\left(\frac{1}{2} \sin^{-1}\left(i \frac{C_3 C_4^* - C_4 C_3^*}{C_3 C_3^* + C_4 C_4^*}\right)/2\right)$$

The Figure of Merit can be calculated from above results as in Equation 7.6

$$FOM = \frac{\theta_F}{\alpha} = \frac{\theta_F d}{-\ln(T)}$$

7.3 A Genetic Algorithm

Genetic Algorithms are global numerical-optimization methods, patterned after the natural processes of mating and mutation\[11-14]\]. Figure 7-2 below is the flowchart of the genetic algorithm developed for 1D PCs.
The basic building blocks in a genetic algorithm are genes. A gene is a binary encoding of a structure of our multilayers. A chromosome is an array of genes. Each chromosome has an associated cost (pattern) function, assigning a relative merit to that
chromosome. A pattern function is associated with both rotation and transmittance of multilayer structures. A simple pattern function can be chosen as the products of transmittance and Faraday rotation angle. The algorithm then begins with a large list of random chromosomes. The process to choose the system merit can be called pattern recognition. The final structure is decided by pattern (cost) functions. The chromosomes are evaluated and ranked from the highest to lowest according to cost functions. Inferior chromosomes are discarded, leaving a superior species or subset, to survive. Surviving genes become parents, and then they swap some of their genetic structure to produce two new offspring. This swap of genetic information is achieved by splitting both parents at a random position in the chromosome, and then swapping tails. Through this process, the parents reproduce enough to replace the discarded chromosomes. Mutations cause small random changes in a chromosome. Mutation rate is set to 10% for new whole offspring. Cost functions are evaluated for new offspring and the mutated chromosome, and the process is repeated until an acceptable solution is obtained or after a preset number of iterations.

How good the result of the aperiodic structure analysis with genetic algorithm is depends mainly on how to evaluate the pattern (cost) function of a structure. Therefore, the pattern recognition is critical to evaluate the transmittance and Faraday rotation angle.

7.4 Simulation Results

Since the calculation is huge, the computation is done with high performance computing techniques and systems. To cooperate with our current research for garnet’s
Faraday rotation in the visible regime, the incident light wavelength is chosen as the Nd:YAG second harmonic at 532nm. The typical dielectric tensor of Bismuth substituted Yttrium Iron Garnet (BYIG) chosen are $\varepsilon_1=6.09 + i 0.05$ and $\varepsilon_2=0.018 + i 0.008$. The tensor $\varepsilon_0$ of dielectric material SiO$_2$ is 2.18. After the genetic algorithm’s optimization, a best structure determined by pattern function is represented $d_{M1}, d_{D1}, \ldots, d_{Di}, \ldots, d_{MN}$.

Then a thickness distribution of the best coating structure is obtained. Figure 7-3 shows a single 2μm magnetic layer (#1) and TWO optimized structures corresponding to 2 pattern functions. Structure #2 is primarily optimized for Faraday rotation angle per magnetic material thickness and secondly for transmittance. Structure #3 is primarily optimized for total Faraday rotation angle and the figure of merit. Structure * is a quarter-wavelength stack for comparison, which is made up of alternately high- and low-index quarter-wave layers.
Table 7-1 lists the Transmittance (T), total rotation angle $\theta$, Figure of Merit (FOM=2$\theta$/\alpha), Faraday rotation per magnetic material thickness ($\theta$/d$_M$) for 2 different structures and a single 2$\mu$m magnetic layer for comparison. The transmittance and Faraday rotation of #1 a single 2$\mu$m BYIG film are 34.31% and 4.11° respectively. #2, and #3 corresponds to the structure listed in Figure 7-3.

<table>
<thead>
<tr>
<th>Structure</th>
<th>Transmittance</th>
<th>$\theta$</th>
<th>FOM</th>
<th>$\theta$/d$_M$</th>
</tr>
</thead>
<tbody>
<tr>
<td>#1</td>
<td>34.31%</td>
<td>4.11°</td>
<td></td>
<td></td>
</tr>
<tr>
<td>#2</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>#3</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>*</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 7-3: Three structures #1: a single 2$\mu$m magnetic layer; #2: a structure primarily optimized for Faraday rotation angle per magnetic material thickness and secondly for transmittance. #3: primarily optimized for total Faraday rotation angle and the figure of merit. * quarter-wavelength stack for comparison only, which is made up of alternately high- and low-index quarter-wave layers.
Table 7-1: Optical and magneto-optical properties of 4 different structures

<table>
<thead>
<tr>
<th></th>
<th>T</th>
<th>$\theta_{\text{total}}$ (°)</th>
<th>FOM (°)</th>
<th>$\theta/d_M$ (°/μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>#1</td>
<td>34.31%</td>
<td>4.11</td>
<td>7.7</td>
<td>2.06</td>
</tr>
<tr>
<td>#2</td>
<td>38.53%</td>
<td>7.72</td>
<td>16.2</td>
<td>27.98</td>
</tr>
<tr>
<td>#3</td>
<td>20.28%</td>
<td>15.13</td>
<td>19.0</td>
<td>11.74</td>
</tr>
</tbody>
</table>

Structure # 2 has best transmittance and Faraday rotation per magnetic material thickness ($\theta/d_M$). The Faraday rotation per magnetic material thickness ($\theta/d_M$) of structure #2 has been enhanced to 13.6 times than a single magnetic layer which is from $2.06^\circ/\mu$m to $27.98^\circ/\mu$m, while structure # 3 has the highest total Faraday rotation ($2\theta/\alpha$) and the best figure of merit which are 3.7 and 2.5 times more than a single magnetic layer. All these results prove the effectiveness of this newly developed optimization technique.

7.5 Summary

The optical and magneto-optical properties of one-dimensional structures could be optimized for an aperiodic multilayer system with a genetic algorithm. Pattern functions defined the merit of 1D-PCs which were used to compare the relative transmittance and Faraday Effect. Theoretical and simulated results show that aperiodic multilayer structured systems composed of Bi-substituted yttrium–iron–garnet films and dielectric SiO2 films could be optimized for the transmittance or Faraday Effect, or both. The analysis results reveal that the MO and transmittance characteristics of the 1D-PC films are almost entirely governed by the degree of localization of light, which can be controlled by varying the aperiodic structure of multiple layers in the films.
During the computation process, it is found that the pattern function directly affects the resultant structures. That is, some of the best structures may be missed. To confirm the theoretical results experimentally, further studies are needed to fabricate the aperiodic multilayer films with sputtering techniques.

### 7.6 References


Chapter 8

Conclusion

The magneto-optical materials with both high Faraday rotation and high transmittance are highly desired in high speed switches, isolators, and visible imaging systems. In this study, we focused on material synthesis, switching test, and theoretical investigations of several magneto-optical thin-films, crystals, and nanoparticles in the visible range of the spectrum. Specifically, (1) new Bi$_3$Fe$_4$Ga$_1$O$_{12}$ (BIGG) garnet films were synthesized that greatly improved transmittance and maintain good Faraday rotation in the visible range of the spectrum; (2) FeBO$_3$ nanoparticles were fabricated to eliminate birefringence and further improved the FeBO$_3$ crystal’s transmittance; (3) new Bi$_{1.8}$Y$_{1.2}$Fe$_4$Ga$_1$O$_{12}$ (BYIGG) nanoparticles were synthesized with better transmittance; (4) aperiodic multiplayer structures were theoretically investigated to improve both Faraday rotation and transmittance; (5) RF circuit board was specially designed, and the response time of circuit board reached our requirements.

Table 8-1 is the summary of the properties of different magneto-optical materials. For comparison, some related materials and the theoretically optimized multi-layer structure are also included.
BIGG films have been successfully synthesized on garnet substrates by a pulsed laser deposition technique. X-ray diffraction and rocking curve analyses confirmed the epitaxial quality of the films with a lattice constant close to 12.61±0.01 Å. Compared with that of a bismuth iron garnet Bi$_3$Fe$_5$O$_{12}$ (BIG) film, the bandwidth of the BIGG transmittance (T) spectrum was extended and its left edge shifted down in wavelength by approximately 50 nm. This enables its Faraday rotation $\theta_F$ and switching speed to be tested at a wavelength of 532nm. While BIGG films had a Faraday rotation angle slightly

<table>
<thead>
<tr>
<th>Materials</th>
<th>Faraday rotation</th>
<th>Figure of merit*</th>
<th>Natural birefringence</th>
<th>Switching speed</th>
<th>Discussed in this thesis</th>
</tr>
</thead>
<tbody>
<tr>
<td>BIGG thin-film</td>
<td>16.2°/μm</td>
<td>9.0°</td>
<td>no</td>
<td>--</td>
<td>chapter 3</td>
</tr>
<tr>
<td>BIGG thin-film</td>
<td>15.6°/μm</td>
<td>16.5°</td>
<td>no</td>
<td>2.4ns</td>
<td></td>
</tr>
<tr>
<td>FeBO$_3$ crystal</td>
<td>0.23°/μm (intrinsic) 1.5°/60μm (measured)</td>
<td>104° (calculated)  6.9° (measured)</td>
<td>yes</td>
<td>1.2ns</td>
<td>chapter 4</td>
</tr>
<tr>
<td>FeBO$_3$ nanoparticles</td>
<td>~0.08°/μm (calculated)</td>
<td>&gt;34.7° (expected)</td>
<td>eliminated</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td>BYIG nanoparticles</td>
<td>0.8~100/mm (reported) 0.2° to 9.6° (reported)</td>
<td>no</td>
<td>--</td>
<td>chapter 5</td>
<td></td>
</tr>
<tr>
<td>BYIGG nanoparticles</td>
<td>2°/mm</td>
<td>2.2°</td>
<td>no</td>
<td>--</td>
<td>chapter 5</td>
</tr>
<tr>
<td>BYIG single layer</td>
<td>2.1°/μm (calculated) 7.7° (calculated)</td>
<td>no</td>
<td>---</td>
<td>chapter 7</td>
<td></td>
</tr>
<tr>
<td>BYIG multi-layer structure</td>
<td>7.5°/μm (optimized) 19.0° (optimized)</td>
<td>no</td>
<td>---</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Figure of merit is defined as $2\theta/\alpha$
-- not tested or --- not applicable (theoretical study only)
less than that of BIG films, their figure of merit have been greatly improved to a typical value about 1.8 times larger than that of the BIG films. Further, the switching time of the BIGG films has reached 2.4 ns with a Φ1 mm circular aperture at the wavelength of 532 nm.

Flux sintering technique was utilized to grow Iron Borate (FeBO₃) crystals. Our test shows that the green platelets we grew had good (001) crystallization. The switching speed of FeBO₃ reached 1.2ns while the figure of merit was measured of 6.9°. FeBO₃ nanoparticles have been successfully synthesized by a high energy ball-milling technique to a thickness of around 30 nm. It is predicted that FeBO₃ nanoparticles have the potential to overcome the birefringence, improve the transmittance, and meantime have similar switching speed as FeBO₃ crystals.

Bi₁₈Y₁₂Fe₄Ga₁₂O₃₂ (BYIGG) nanoparticles have been successfully fabricated with coprecipitation and annealing processes, where gallium was doped to improve the transmittance of the films. With a high energy ball-milling technique, BYIGG films were coat-spun on glass substrates. A reasonable Faraday rotation of ~ 2.5° was obtained with 0.5mm thick films, and the transmittance of these films at 532nm was of about 1%. The figure of merit (2θ/α) was found to be ~ 2.2°, which is among the reported range between 0.2° and 9.6°. The possible reason for the lower figure of merit is the contamination, which needs more experiments and investigation to verify.

In addition, we proposed a new optimization technique and successfully obtained aperiodic multilayer structures with both enhanced Faraday effect and better transmittance. One-dimensional magnetic crystals of aperiodic multilayer structures were
theoretically studied and optimized by means of a genetic algorithm. Bi-substituted yttrium–iron–garnet films and dielectric SiO₂ films were used in the simulation at the wavelength of 532nm. The Faraday rotation per magnetic material thickness (θ/dₘ) of one structure has been enhanced from 2.06°/μm to 27.98°/μm, which is 13.6 times that of a single magnetic layer. The other optimized structure has a higher total rotation angle (3.7 times) and the better figure of merit (2.5 times) than a single magnetic layer. All these results proved the effectiveness of this newly developed optimization technique.

Circuit board and coil design are critical for switching speed test. If the circuit response time is larger than the films/crystals’ response times, the speed tested is limited by the driving circuit board. Therefore, a RF circuit and a miniature coil were specially designed to generate a magnetic field about 400 Oe. A circuit rise time less than 1 ns was reached.

Future studies will focus on following aspects: (1) optimize BIGG thin films and grow more garnet films with other dopants; (2) finish synthesizing FeBO₃ nanoparticle films and testing; (3) decrease the contamination of BYIGG nanoparticle films, and increase their figure of merit; (4) sputter aperiodic thin-films to improve transmittance and rotation according to our theoretical derivation.
Appendix

Determination of the thickness and the refractive index of garnet thin-films

The thickness of thin film on a transparent substrate can be estimated from transmittance interference fringes base on multiple reflections at the three interfaces as shown in Figure H-1 \cite{1}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure_h1.png}
\caption{The structure of a thin-film on a thick transparent substrate}
\end{figure}

The transmittance of a Russia garnet film(#110) was measured as shown in Figure H-2.
The substrate of Russia sample is confirmed as GGG (111). The known refractive index is 2.02 \(^2\). For GGG substrate, the interference-free transmittance is found to be 79.52% from Equation H.1

\[
T_s = \frac{2s}{s^2 + 1}
\]  

Eq. H.1

First, the transmittance of our sample is calibrated with Ts. Second, we find the local minimum and maximum as shown in Figure H-3.
Third, an envelope is interpolated based on these local minimum and maximum as shown in Figure H-4.

Figure H-3: The local minimum and maximum of the transmittance spectrum
In weak and medium absorption region, the refractive index is estimated as a function of wavelength from Equation H.2

$$n = \left[ N + \left( N^2 - s^2 \right)^{1/2} \right]^{1/2}$$

$$N = 2s \frac{T_M - T_m}{T_M T_m} + \frac{s^2 + 1}{2}$$

Eq. H.2

where $T_M$ and $T_m$ are interpolated maximum and minimum, respectively.

Forth, a least-square fit $n(\lambda) = a/\lambda^2 + c$ is used for the garnet material and we find $a=4.3568 \times 10^4$ and $c=2.2924$; the fitting refractive index curve verses wavelength is shown as in Figure H-5.
The thickness can be found at minimum and maximum points according to Equation H.3

\[ d = \frac{\lambda_1 \lambda_2}{2(\lambda_1 n_2 - \lambda_2 n_1)} \]  

Eq. H.3

Then we got 10 preliminary thickness results in Table H-1

Table H-1: 10 preliminary thickness results

<p>| | | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>2.7047</td>
<td>2.7569</td>
<td>2.4355</td>
<td>2.6517</td>
<td>2.4355</td>
<td></td>
</tr>
<tr>
<td>2.8186</td>
<td>2.6388</td>
<td>2.6306</td>
<td>2.5871</td>
<td>2.5343</td>
<td></td>
</tr>
</tbody>
</table>

Figure H-5: The fitting refractive index curve verses wavelength
These values are used to determine the orders of interference fringe. The orders of the interference fringes is calculated from Equation H.4

\[ 2nd = m\lambda \]

where \( m \) is the order of fringes that is either an integer or a half integer. The \( m \) calculated is close to an integer for maximum and a half integer for a minimum point. When \( m \) is replaced with a corresponding integers or half-integers, recalculated \( d \) has more accurate result. The \( m \) value calculated and modified is listed in Table H-2

<table>
<thead>
<tr>
<th>the order ( m ) (calculated)</th>
<th>the order ( (m) ) (modified)</th>
</tr>
</thead>
<tbody>
<tr>
<td>21.59 20.62 19.67 18.59 17.60</td>
<td>21.5 20.5 19.5 18.5 17.5</td>
</tr>
<tr>
<td>22.01 21.07 20.08 19.08 18.07</td>
<td>22 21 20 19 18</td>
</tr>
</tbody>
</table>

The order of fringes \( m \) is then substitute back into formula 4, a more accurate thickness is found to be \( 2.6063\mu m \)

Reference:

2. [http://socrates.berkeley.edu/~eps2/wisc/ri.html](http://socrates.berkeley.edu/~eps2/wisc/ri.html)
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- Shaoying Kang, Shizhuo Yin, Qi Li, Venimadav Adyam, and Yong Zhu, “Bi$_2$Fe$_4$Ga$_{12}$O$_{32}$ garnet properties and its application to ultrafast switching in visible”, IEEE trans. On Magnetics, accepted
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