Optical properties of gyrotrropic polygonal chiral thin films

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Abstract
The effect of gyrotrropy on optical response of dielectric polygonal chiral thin film was reported using transfer matrix method. The influence of gyrotrropy was studied on selective transmission and reflection spectra for polygonal chiral thin films at different void volume fractions and oblique incident angles of light. The results revealed that the Bragg regimes and outside Bragg regimes of polygonal chiral thin films were affected by high induced gyrotrropy.

Keywords Chiral thin films · Gyrotropy · Bragg regime

1 Introduction

Magneto-optical materials in a magnetic field exhibit a non-reciprocity property is called gyrotrropic effect (Boardman and Xie 2003). Even in the real life, magneto-optical materials can reveal the nonreciprocal properties without magnetic field, for example in ferromagnets (Mansuripur 1995; Zvezdin and Kotov 1997). The optical characteristics of non-homogenous and anisotropic thin films with gyrotrropy property are same as plasmas and ferrites (Chen 1983). There are three kinds of gyrotrropy in the names of free, forced and natural. Free gyrotrropy is related to internal magnetic fields and forced gyrotrropy is associated with external magnetic fields. In a natural gyrotrropy, the rotation of plane of polarization is reversible (e.g. sugar solution) (Boardman and Xie 2003). A gyrotrropic medium can make a difference between left-handed circular polarization (LCP) and right-handed circular polarization (RCP) of light.

Polygonal chiral thin films are three dimensional artificial nanostructures as a stack of biaxial plates that can be produced by combination of oblique angle deposition technique and abrupt changes of substrate rotation about its substrate normal (Hodgkinson et al. 2004; Popta et al. 2005). A polygonal chiral thin film can be formed with the discrete abrupt angular rotations of $2\pi/Q$, where the parameter $Q > 2$ is an integer. A polygonal chiral thin film with $Q = 4$ is called equichiral sculptured thin film (or tetragonal) and other is named ambichiral sculptured thin film (Mackay et al. 2013).
Chiral sculptured thin films are local biaxial (Lakhtakia and Messier 2005; Lakhtakia 2000) and there is a selection in reflection RCP and LCP from them in Bragg regime, which is known as circular Bragg phenomenon (CBP) (Faryad and Lakhtakia 2014). The effect of gyrotropy on the CBP exhibited by chiral sculptured thin films (Q > 10) (Pickett et al. 2004; Pickett and Lakhtakia 2002). In magneto-optical materials, the off-diagonal components of the relative permittivity tensor can be activated (this property is called gyrotropy) (Eroglu 2010). Then, the mutuality between circular Bragg phenomenon and gyrotropy effect could be interested in optics of chiral magneto-optic thin films.

In this work, the problem of light propagation with circular polarization was investigated in gyrotropic polygonal chiral thin film. The effect of gyrotropy on polygonal chiral thin films at different void volume fractions and oblique incident angles of light was investigated. The theory in brief is presented in Sect. 2 for optical modeling and followed by results and discussion in Sect. 3.

1.1 Theory in brief

Consider the region $0 \leq z \leq d$ is occupied by a gyrotropic polygonal chiral thin film, while the regions $z \leq 0$ and $z \geq 0$ are vacuous (Fig. 1). Let us assume that this structure can be exposed by a circular polarized plane wave from the bottom of the structure at an angle $\theta$ to the $z$-axis. The phasors of incident, reflected and transmitted electric fields are given:

$$
E_{\text{inc}}(r) = \left[ \frac{(-S+P_z)}{\sqrt{2}} a_L - \frac{(iS+P_z)}{\sqrt{2}} a_R \right] e^{i k(z \cos \theta + x \sin \theta)}, \quad z \leq 0
$$

$$
E_{\text{ref}}(r) = \left[ \frac{(S-P_z)}{\sqrt{2}} r_L + \frac{(iS+P_z)}{\sqrt{2}} r_R \right] e^{i k(-z \cos \theta + x \sin \theta)}, \quad z \leq 0
$$

$$
E_{\text{tr}}(r) = \left[ \frac{(iS-P_z)}{\sqrt{2}} t_L - \frac{(iS+P_z)}{\sqrt{2}} t_R \right] e^{i k((z-d) \cos \theta + x \sin \theta)}, \quad z \geq d
$$

The phasor of the magnetic field in any region is given as $H(r) = (i \omega \mu_0)^{-1} \nabla \times E(r)$, where $(a_L, a_R), (r_L, r_R)$ and $(t_L, t_R)$ are the amplitudes of incident plane wave, and reflected and transmitted waves with LCP and RCP. The $S$ and $P_z$ are respectively unit vectors for linear polarization normal and parallel to the incident plane and also $u_{x,y,z}$ are Cartesian unit vectors. The reflectance and transmittance amplitudes can be obtained, using the continuity
of the tangential components of electrical and magnetic fields at two interfaces of structure and solving the algebraic matrix equation (Lakhtakia and Messier 2005):

\[
\begin{bmatrix}
    i(t_L - t_R) \\
    -(t_L + t_R) \\
    0 \\
    0
\end{bmatrix}
\begin{bmatrix}
    K(\theta_0) \end{bmatrix}^{-1}
\begin{bmatrix}
    [M_p] \\
    [K(\theta_0)]
\end{bmatrix} =
\begin{bmatrix}
    i(a_L - a_R) \\
    (a_L + a_R) \\
    -i(r_L - r_R) \\
    (r_L + r_R)
\end{bmatrix}
\]

(2)

where the different terms and parameters of this equation are given in detail in Ref (Lakhtakia and Messier 2005). Finally, using the obtained amplitudes in Eq. 2, then we can calculate the reflectance and transmittance coefficients as

\[
\begin{align*}
    r_{ij} &= \frac{r_j}{a_i}, \\
    t_{ij} &= \frac{t_j}{a_i}; \quad i,j = L,R.
\end{align*}
\]

The reflection and transmission is calculated as

\[
\begin{align*}
    R_{ij} &= \left| \frac{r_{ij}}{a_i} \right|^2, \\
    T_{ij} &= \left| \frac{t_{ij}}{a_i} \right|^2; \quad i,j = L,R.
\end{align*}
\]

The nonhomogeneous dielectric permittivity \( \varepsilon_p \) for \( l \) th arm of the polygonal chiral is defined as (Mackay et al. 2013):

\[
\varepsilon_p = \sum_{i,j} S_i(\zeta)S_j(\chi)\varepsilon_{\text{ref}} S_{ij}^{T}(\chi)S_{ij}^{T}(\zeta)
\]

(3)

where the superscript \( T \) indicates to the transpose of a dyadic, \( \zeta = h(l-1) (2\pi/Q) \), \( \chi \) is tilt angle of the nanocolumns of the polygonal chiral, \( h = +1(\text{or}-1) \) is right-handedness (or left-handedness) of the polygonal chiral. In our work, \( Q \) was fixed as 3 (trigonal),4 (tetragonal),5 (pentagonal),6 (hexagonal) and 12 (dodecagonal).

The local relative permittivity, rotation and tilt dyadics are respectively as (Pickett et al. 2004; Pickett and Lakhtakia 2002):

\[
\begin{align*}
    \varepsilon_{\text{ref}} &= \varepsilon_{\text{a},u_xu_x} + \varepsilon_{\text{b},u_xu_y} + \varepsilon_{\text{c},u_yu_z} + i\varepsilon_{\text{g}} (u_xu_y - u_yu_x) \\
    S_z &= (u_xu_x + u_yu_y) \cos \zeta + (u_xu_y - u_yu_x) \sin \zeta + u_zu_z \\
    S_y &= (u_xu_x - u_yu_y) \cos \chi + (u_xu_y + u_yu_x) \sin \chi + u_zu_z
\end{align*}
\]

(4)

where \( \varepsilon_{\text{a,b,c}} \) are the relative permittivity scalars and \( \varepsilon_{\text{g}} \) indicates to gyrotropy surface of polygonal chiral in \( yz \)-plane.

\section{2 Results and discussion}

In order to optical modeling, at first we considered a structure as a right-handed (\( h=+1 \)) TiO\(_2\) porous polygonal chiral thin film (Fig. 1). In optical modeling, the relative permittivity scalars \( \varepsilon_{\text{a,b,c}} \) in Eq. 4 are calculated using the Bruggeman homogenization formalism (Sherwin and Lakhtakia 2002, 2003). In this formalism, the structure is considered as a two-component composite (TiO\(_2\) and void). These quantities are dependent on different parameters, namely columnar form factor, fraction of void (\( f_v \)), the wavelength of free space, and the refractive index. In addition, each column in the structure is considered as a string of identical long ellipsoids. The ellipsoids are considered to be electrically small (i.e., small in a sense that their electrical interaction can be ignored) (Lakhtakia and
Messier 2005). In all calculations, columnar form factors were fixed as \((c/a)_{\text{TiO}_2, \text{void}} = 20\) and \((b/a)_{\text{TiO}_2, \text{void}} = 1.06\) (Sherwin et al. 2002) (c, a & b are the semimajor axis and small half-axes of ellipsoids, respectively). We were used the frequency-dependent bulk experimental dielectric function of \(\text{TiO}_2\) (Palik 1985) for homogenization. The selective transmission (ST) and selective reflection (SR) is calculated as \(\text{ST} = T_{LL} - T_{RR}\) and \(\text{SR} = R_{LL} - R_{RR}\), respectively.

It must be mentioned here, we supposed a static magnetic field in \(x\)-direction \(H = H_0 u_z\). In simplest case, to first order a gyration vector \(g = (e_x, 0, 0)\) is proportional to the applied magnetic field, where \(\epsilon_g = \epsilon_0 \chi^{(m)} H_0\) (Eroglu 2010) and while \(\chi^{(m)}\) is the magneto-optical susceptibility and it was a scalar and frequency independent in our simulation. In generally, it can be frequency dependent due to dispersion of \(\chi^{(m)}\) and even can be a tensor. In most cases, the gyrotyropy factor is considered as \(\epsilon_g = \epsilon_0 \left(\omega_p^2 \omega_c^2 / \omega^2 (\omega^2 - \omega_c^2)\right)\), where \(\omega_p = \sqrt{Ne^2 / \epsilon_\infty m_{\text{eff}} \omega_c = e \mu_0 H_0 / m_{\text{eff}}}\) are the plasma and cyclotron frequencies respectively, \(\epsilon_\infty\) is the background permittivity, \(N\) is the electron density, \(m_{\text{eff}}\) is the effective mass, and \(e\) is the electron charge (Mansuripur 1995; Zvezdin and Kotov 1997). Therefore, the effect of gyrotyropy in structure was considered as above not as a porous polygonal chiral thin film infiltrated with magnetic nanoparticles or a porous polygonal chiral thin film fabricated from magnetic materials. Also, the azimuthal angle of incident light was \(0^\circ\), the tilt angle of nanocolumns, the structural period (pitch) and the thickness of polygonal chiral thin film respectively were \(42^\circ\), \(2\Omega = 325 \text{ nm}\) and \(d = 3\Omega\).

Selective transmission from right-handed \((h = +1)\) \(\text{TiO}_2\) polygonal chiral thin films (non-gyrotropic) for types of polygon shapes at different void volume fractions with \(\theta = 0^\circ\) is shown in Fig. 2. In our work, there exist two Bragg resonances (two Bragg regimes) due to fixing the thickness and pitch of polygonal chiral thin films, although by selection of suitable structural parameters for biaxial plates in chiral sculptured thin films can occur the multiple Bragg regimes. This work is in agreement with the experimental work of Lakhtakia’s group in the number of Bragg regimes in a certain condition with \(\phi = \Delta \zeta\) (the rotation angle between the arms of polygonal chiral thin film that it is an incremental angle in Ref (Hodgkinson et al. 2004)). For example, a tetragonal chiral thin film with \(\phi = 90^\circ\) in our work is the same as the equichiral \((p = 0, q = 2)\) in Ref (Hodgkinson et al. 2004). In this work, first and second Bragg resonances are \(\lambda^{(1)}_{BR}\) and \(\lambda^{(2)}_{BR}\), respectively, equal to \(G^{(1)}(0, 0) = 1\) and \(G^{(2)}(0, q) = q - 1\) from Lakhtakia’s group (Hodgkinson et al. 2004). On the other hand, \(G^{(1)}(0, 0) / G^{(2)}(0, q) = 1 / q - 1\) is the same as \(\lambda^{(1)}_{BR} / \lambda^{(2)}_{BR} \approx 1 / (180^\circ / \phi)\) - 1 so that \(180^\circ / \phi\) is defined as parameter \(q\) in Ref (Hodgkinson et al. 2004). The first Bragg resonance (BR) centered around at wavelength \(\lambda^{(1)}_{BR} \approx 2n_{\text{avg}} \Omega\), where \(n_{\text{avg}}\) is average refractive index of polygonal chiral thin film and the second Bragg resonance almost located at wavelength \(\lambda^{(2)}_{BR} \approx (2 / (Q - 2)) \lambda^{(1)}_{BR}\) where \(Q = 360^\circ / \phi\). It is well known that when \(\epsilon_g = 0\), the average refractive index of porous polygonal chiral thin film is \(n_{\text{avg}} = \sqrt{\epsilon_x + \sqrt{\epsilon_x^2 + \epsilon_y^2}}\) \(\cos^{1/2} \theta\), where \(\epsilon_d = \frac{\epsilon_x + \epsilon_y}{\epsilon_x \cos^2 \theta + \epsilon_y \sin^2 \theta}\) is composite relative permittivity scalar (Lakhtakia and Messier 2005). If we consider \(\epsilon_g \neq 0\), the relative permittivity scalars \(\epsilon_{a,b,c}\) are modified as \(\epsilon_b \rightarrow \epsilon_b + \epsilon_g\)

\[\epsilon_c \rightarrow \frac{(\epsilon_c + \epsilon_g) + \sqrt{(\epsilon_c - \epsilon_g)^2 + 4\epsilon_g^2}}{2}\]  

and \(\epsilon_a \rightarrow \frac{(\epsilon_a + \epsilon_g) - \sqrt{(\epsilon_a - \epsilon_g)^2 + 4\epsilon_g^2}}{2}\). These new relative permittivity scalars can be obtained from diagonalization the local relative permittivity in Eq. 4. It is enough that the new relative permittivity scalars to be replaced in definition of the average refractive index. It is found that the intensity and width of Bragg regimes increased by increasing the porosity of polygonal chiral thin films from 0.2 to 0.8. On the other hand,
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increase porosity of polygonal chiral thin film enhances the circular Bragg phenomenon (in intensity and width). In all types of polygon shapes, the Bragg regimes shift to shorter wavelengths with increase of porosity. It can be seen that the optical properties of polygonal chiral thin films are same as the chiral sculptured thin films at polygon shapes with more vertices. For $Q = 4$ (tetragonal), two Bragg resonances coincide at wavelength $\lambda^{BR}$ for different porosity of polygonal chiral thin films. Also, the calculations were performed for oblique incident angle of light and obtained same results. Here, only for $Q = 3$ (trigonal shape) density plots as functions of wavelength and incident angle $[0°–45°]$ at different void volume fractions depicted in Fig. 3, while other parameters are same as Fig. 2. It is clear that in Fig. 3 the both Bragg regimes move to shorter wavelengths with increasing porosity of structure.

The calculations of selective reflection & transmission spectra were performed for all polygonal shapes at different void volume fractions and oblique incident angles with values of the level gyrotropy at $-5 \times 10^{-6}$ and $+5 \times 10^{-6}$ (weak induced gyrotropy). The obtained results showed that the weak induced gyrotropy had not significant effect on co- and cross-handed reflection and transmission spectra, are not shown here. Also, we found that a gyrotropy parameter that is smaller by four (or more) orders of magnitude of $\varepsilon_{a,b,c}$ is not going to effect on the Bragg regimes and outside Bragg regimes. The oscillations outside of Bragg regime can be related to the Fabry–Perot oscillations (Mahalik 2006). But, the largest magnitude of $\left|\varepsilon_{a}\right|$ for an actual material by Levy and Karki was reported 0.05 (Levy and

![Fig. 2 Selective transmission from right- handed(h = + 1) non-gyrotropic TiO$_2$ polygonal chiral thin films at different void volume fractions with $\theta = 0°, 2\Omega = 325$ nm, $d_{RP} = 3\Omega$ and $\gamma = 42°$. Inset plots show a single column of polygonal chiral thin films and its cross section](image-url)
Fig. 3 Density plots of right-handed non-gyrotropic TiO$_2$ trigonal chiral thin film (Q=3) as functions of wavelength and incident angle at different void volume fractions.

Fig. 4 Selective transmission & reflection of hexagonal chiral thin film (Q=6) with $f_v = 0.4$ for different gyrotropies at $\theta = 0^\circ$, $\theta = 30^\circ$. 
Karki 2017). Anyway, in order to influence of gyrotnopy on selective transmission and reflection, we considered the value of gyrotnopy from six orders smaller to one order than $\varepsilon_{a,b,c}$, is shown in Fig. 4 for hexagonal chiral thin film ($Q=6$) with $f_v=0.4$ at $\theta = 0^\circ$, $\theta = 30^\circ$. It is clear that the ST and SR spectra for gyrotnropic polygonal chiral thin film are same as non-gyrotnropic polygonal chiral thin film at low gyrotnopies but the Bragg regimes and outside Bragg regimes of spectra affected at high gyrotnopies ($\varepsilon_g$ s two orders or one order smaller than $\varepsilon_{a,b,c}$). Then, in the following in Figs. 5, 6, 7, 8 and 9 we only considered $\varepsilon_g$ two orders smaller ($\varepsilon_g = +5 \times 10^{-2}$) than $\varepsilon_{a,b,c}$.

The influence of gyrotnopy on selective transmission & reflection spectra for different polygonal chiral thin films with $f_v=0.4$ and $\theta = 0^\circ$ is given in Fig. 5. A comparison between obtained results showed that the spectra had changes in the Bragg regimes and outside Bragg regimes at $\varepsilon_g = +5 \times 10^{-2}$ for different polygonal chiral thin films to those obtained at $\varepsilon_g = 0$(although these changes was slight in Bragg regimes). The induced gyrotnopy is created the oscillations in spectra due to activation the off- diagonal components of the relative permittivity tensor. It is observed that the oscillations in outside Bragg regimes (Fabry–Perot oscillations) are more than Bragg regimes. It is well known that the effect of gyrotnopy for $Q>10$ is same as sculptured chiral thin film. We repeated calculations for trigonal chiral thin film ($Q=3$) in Fig. 6 at different void volume fractions. It is found that the obtained results are same as Fig. 5. Although the created oscillations by gyrotnopy decreased from 0.2 to 0.8 due to decrease in the density of the polygonal chiral thin film. A comparison for ST and SR spectra in Figs. 5 and 6 showed these oscillations do not exist in non-gyrotnropic polygonal chiral thin film. Then it can be concluded that the gyrotnopy property created them in polygonal chiral thin films due to nonzero the off- diagonal components of the relative permittivity tensor. A comparison between positive and negative values of gyrotnopy in ST and SR spectra for trigonal chiral thin film ($Q=3$) with $f_v=0.4$ at $\theta = 0^\circ$, $\theta = 30^\circ$ is shown in Fig. 7. It is seen that the first and second Bragg regimes got wider at oblique incident angle of light. The plots revealed that the oscillations in outside Bragg regimes do not match at positive and negative values of gyrotnopy. Even the Bragg regions shift left or right depending on the positive and negative values due to changing direction of magnetic static field.

Density plots of selective transmission and reflection of trigonal chiral thin film ($Q=3$) for normal incident of light ($\theta = 0^\circ$) as functions of wavelength and gyrotnopy at different void volume fractions are depicted in Fig. 8. The obtained results showed that the both of Bragg regimes shift to shorter wavelengths with increasing porosity of polygonal chiral thin film. Oscillations in and outside Bragg regimes in density plots of selective transmission still exist due to variations of refractive index of polygonal chiral thin film. It is found that the intensity of Fabry–Perot oscillations decreased in selective transmission with increasing of porosity of polygonal chiral thin film from 0.2 to 0.8. Because the optical constants of structure get closer to vacuous medium.

The calculations were repeated for oblique incident angles of light with $f_v=0.4$ that they are given in Fig. 9. The obtained results are same as Fig. 8. The only difference was between selective transmission and reflection spectra in direct and inverse Bragg regimes. The chiral sculptured thin films almost completely reflect the circular polarization state of normally incident light in the Bragg regime when it matches the structural handedness of the material, while the opposite circular polarization state can freely propagate (Popta et al. 2005). A polygonal chiral thin film can be described as a stack of biaxial plates that rotate by $2\pi/Q$ increments in the clockwise direction as seen by RCP light and by $\pi(Q−2)/Q$ increments in the counterclockwise direction as seen by LCP light (Hodgkinson et al. 2004; Popta et al. 2005); these different rotation rates
Fig. 5 The influence of gyrotropy on selective transmission & reflection spectra for different polygonal chiral thin films with \( f_\gamma = 0.4 \) and \( \theta = 0^\circ \)
Fig. 6 The influence of gyrotropy on selective reflection & transmission spectra for different void volume fractions with $Q = 3$ and $\theta = 0^\circ$. 
are responsible for the difference Bragg regimes in transmission and reflection spectra. Obtained results in Figs. 8 and 9 showed that the width of Bragg resonances in density plots are symmetric to the $\varepsilon_g = 0$ for polygonal chiral thin film at $\theta = 0^\circ$, while it is antisymmetric at oblique incident of light. It is found that the both Bragg regimes became wider with increasing incident angle of light and also the oscillations increased in spectra.

3 Conclusions

The optical properties of gyrotropic polygonal chiral thin films were investigated using transfer matrix method. It is found that the optical response of gyrotropic polygonal chiral thin films at weak induced gyrotropy (in our work $|\varepsilon_g| \in [10^{-6}, 10^{-4}]$) is same as non-gyrotropic polygonal chiral thin films. It is obtained that the selective transmission and reflection spectra for gyrotropic polygonal chiral thin films were affected by strong induced gyrotropy (in our work $|\varepsilon_g| \approx 10^{-2}$). The strong induced gyrotropy is created oscillations in Bragg regimes and outside Bragg regimes due to activation the off-diagonal components of the relative permittivity tensor. The intensity of created oscillations by gyrotropy decreased when the porosity of chiral thin film increased from 0.2 to 0.8. The obtained results showed that the first and second Bragg regimes got wider at oblique incident angle of light. We found that the selective transmission and reflection spectra in Bragg regimes and outside Bragg regimes do not match at high positive and negative values of gyrotropy.
Fig. 8 Density plots of selective transmission and reflection of trigonal chiral thin film (Q = 3) as functions of $\lambda$ and $\varepsilon_g$ for different void volume fractions at $\theta = 0^\circ$. 
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Fig. 9  Density plots of selective transmission and reflection of trigonal chiral thin film (Q = 3) as functions of \( \lambda \) and \( \varepsilon_g \) at different oblique incident angles with \( f_r = 0.4 \).
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