Multi-mode circular dichroism in n-fold rotational symmetric metamaterials

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Abstract
In this article, the effects of the rotation angle between upper and lower n-fold rotational symmetric nano-structures are studied. Various modes of circular dichroism (single wavelength band, dual-wavelength bands, and more than two wavelength bands) in the wavelength band from 3.3 to 5 μm are realized. Circular dichroism up to 0.798 are observed. Meanwhile, sensitivity of circular dichroism to the rotation angle and reconfigure strategy for opposite responses has been discussed. Based on Born-Kuhn model, physical mechanism of mode’s switching is explained with charge distributions. The multi-mode chiroptical responses in mid-infrared band and the variety of design strategies have potential applications in the field of tunable multi-band chiral devices.

Keywords Chiral metamaterial · Differential transmittance · Circular dichroism · Multi-mode · Absorption

1 Introduction

Metamaterial is an artificial material with many electromagnetic properties that do not exist in nature, such as metamaterial absorber (Landy, et al. 2008; Liu, et al. 2010; Chen, et al. 2021; Zhang, et al. 2020), negative refraction (Valentine, et al. 2008), electromagnetically induced transparency-like (Xu, et al. 2019). It has provided excellent solutions for modulators, sensors, etc. in recent decades (Bu, et al. 2016; Chen, et al. 2020). When metamaterials have chiral properties, such as giant optical activity; circular dichroism; negative refractive index and etc. (Li, et al. 2013, 2014, 2020; Cheng, et al. 2016; Hannam, et al. 2014), they can be called chiral metamaterials. Chiral metamaterials have significant improvement in chiral parameters compared with natural chiral materials because of their strong electromagnetic coupling. Therefore, chiral metamaterials have been widely used to enhance chiral signals in molecular analysis, bio-sensor, chiral signal switch, or occasions with chiral signals (Ma, et al. 2017; Yoo and Park 2019). As a typical phenomenon of
chirality, different responses to left-hand circularly polarized (LCP) waves and right-hand circularly polarized (LCP) waves have been widely utilized in protein structure examining, nanostructural analyzing, medicine detecting, etc. (Berova, et al. 2000; Ranjbar and Gill 2009).

Bi-layer structures are often used in the studies of chiral metamaterials (Kaya 2014; Yan, et al. 2017; Ma, et al. 2016; Kaya, et al. 2017; Zhou, et al. 2012). Some of those works replacing the material of the dielectric layers(Kaya, et al. 2017; Yan, et al. 2017; Zhou, et al. 2012), changing some geometric parameters of the structures(Ma, et al. 2016; Kaya, et al. 2017; Liu, et al. 2019), or adjusting the incident angle of circularly polarized waves(Kaya 2014) in order to modulate the responses of metamaterial to circularly polarized waves. As far as we know, in similar metamaterials, there are only a few papers (Wu, et al. 2013; Liang, et al. 2021; Asgari, et al. 2020) have discussed the relative angle between the upper and lower layers. In this paper, relative rotation angles between two layers are able to realize various differential transmittance (ΔT) and circular dichroism (CD) without changing the structure’s size and material properties has been further studied. And by using specific reconfiguration methods, the polarization direction of the response and the sensitivity of the responses to the rotation angle can be adjusted. It’s worth noting that the CD is differential absorption between LCP and RCP waves. So, the CD can be directly provided by measurements of ΔT between LCP and RCP waves when the metamaterials’ reflectance of both LCP and RCP has the same value.

In this article, the effect of the rotation angle on different rotational symmetric metamaterials has been studied in mid-infrared region. As a result, multi-mode ΔT responses; reconfigure strategy for opposite ΔT responses; sensitivity of the ΔT spectra to the rotation angle; and selective absorption are realized. Because of the atmospheric transparent windows, molecular vibration fingerprints, and attractive plasma resonances in mid-infrared region (Stanley 2012; Yin, et al. 2015), this work will benefit the designing of n-fold chiroptical devices and has potential applications in multimode circular polarized wave modulation.

2 Design and simulation

Firstly, the five-fold rotational symmetric metamaterial (Type N5) is illustrated in Fig. 1a–c as example to present the design of these metamaterials. Every unit of the metamaterial, marked by dotted box in the figure, is composed of a silicon dioxide substrate and two vertically placed multiple rotational symmetric gold nano-structures. A polymer film is inserted between those two nano-structures. The entire unit is periodically expanded along the x and y directions with the same period of \( p \). The front and side views of the unit are shown in Fig. 1b and Fig. 1c respectively. As shown in Fig. 1b, the rotation angle of the upper nano-structure is \( \theta \) (counterclockwise). The length of the blades is \( l_2 \) and the side length of the center pentagon is \( l_1 \). The line width of the nano-structures is \( w \). As shown in Fig. 1c, the thickness of polymer film, silicon dioxide substrate, and nano-structures are \( h_1 \), \( h_2 \), and \( h_3 \) respectively. As shown in Fig. 1d, different types and different fold of rotational symmetric units which will be discussed next are illustrated, and some parameters are unchanged as Type N5: line width; thickness of every material; period of units; length of the blades; and the perimeter of the center polygons. In addition, two reconfigure strategies for opposite responses and more flexible designs are marked by dashed lines in Fig. 1d.
The n-fold rotational symmetry refers to the symmetry that the structure completely coincides with itself after the structure rotates $360º/n$ around the axis of rotation. It should be noted that the rotational symmetry mentioned in this paper refers to each layer of gold structure in the unit (as shown in Fig. 1b), not the whole metamaterial after periodic expansion (as shown in Fig. 1a).

In this paper, the metamaterials are designed and optimized by CST Microwave Studio. The considered electromagnetics numerical simulation is finite element method (FEM) in frequency domain, and the type of mesh is tetrahedral. The boundary condition is set to unit cell and the incident waves are set to circular polarization. The direction of incidence is opposite to the z-axis. The refractive index (RI) of polymer film is set to 1.35 (PTFE) (Saadeldin, et al. 2019). The optical constants of gold have been set from Palik’s data (Palik 1998), and the fitted curves are generated by the second-order fitting model in the CST software, as shown in Fig. 2. The silicon dioxide has been set to the lossy type with dielectric constant of 3.9 and loss tangent of 0.025(Ma, et al. 2013). Table 1.

### 3 Results and analysis

In order to discuss the modulation effect of the rotation angle on the transmission spectra more clearly, Type N4 with rotation angle of 0°, 20°, 42°, and 80° are demonstrated in Fig. 3. Here, $t_{RR}$ and $t_{LL}$ represent the co-polarized transmission coefficient of RCP and LCP waves respectively. And $t_{LR}$ and $t_{RL}$ represent the cross-polarized transmission coefficient of RCP and LCP waves respectively. When the rotation angle is set to 0° (i.e., Fig. 3a), significant asymmetric transmission can be observed in middle wavelength band (3.84–4.23 μm) and long wavelength band (4.23–5 μm). When the rotation angle is set to 20° (Fig. 3b), a large asymmetric transmission can be observed in the short wavelength band (3.33–3.84 μm) and long wavelength band. In order to obtain a mode closer to the single-band response, rotation angle of 42° has been chosen to discuss. When the rotation...
The angle is set to 42° (Fig. 3c), large asymmetric transmission can be observed in middle wavelength band, with a relatively small response between the middle and long wavelength bands. And when the rotation angle is set to 80° (Fig. 3d), asymmetric transmission can be seen in short, middle, and long wavelength bands. Also, it can be found that when the

Table 1 Optimized structural parameters in Fig. 1

| Parameters | $l_1$ | $l_2$ | $w$ | $p$ | $h_1$ | $h_2$ | $h_3$ | Gap |
|------------|-------|-------|-----|-----|-------|-------|-------|-----|
| Length(μm)| 1.2   | 1     | 0.2 | 4   | 1.2   | 0.1   | 0.2   | 1   |
wavelength is less than 3.6 μm, there is nearly no asymmetric transmission in this structure. It should be noted that the reflectivity of RCP and LCP waves almost have the same value in Type N4, which is not shown in Fig. 3.

To further investigate the modulation effect of rotation angle on chirality, the ΔT has been assessed by the $\Delta T = |t_{RR}|^2 - |t_{LL}|^2$. As much as 0.71, 0.798, and 0.657 of the ΔT can be reached in three wavelength bands respectively. The ΔT spectra of four different rotation angles are shown in Fig. 3e, where four different combinations of ΔT spectra are marked as Mode 1, Mode 2, Mode 3, and Mode 4. Since the rotation period of the proposed metamaterial is 90°, and in order to fully discover the impact of rotation angle on ΔT spectra, the corresponding change of ΔT spectra under the rotation angle from 0° to 90° have been simulated in Fig. 3f, where the four modes mentioned before are marked by dash lines in the figure.

As shown in Fig. 4, different folds of rotational symmetric structures are further studied. ΔT spectra of three-fold rotational symmetric nano-structure can be modulated with the changing of rotation angle, which is same as Type N4. Five-fold rotational symmetry, as shown as Type N5, it can be observed that the ΔT spectra in the middle wavelength band can be modulated by rotation angle which is like Type N3 and Type N4. However, the ΔT responses in long wavelength band will not be modulated clearly. To further investigate the relationship between the fold of rotational symmetry and ΔT responses, the ΔT spectra of the nano-structures with six and eight folds of rotational symmetry have been simulated, as shown in Fig. 4c and d, where the ΔT spectra are almost unchanged with different rotation angles.

Fig. 4 a, b, c, and d are the ΔT spectra of the metamaterial with three, five, six, and eight folds of rotational symmetry respectively. The rotation angle of Type N3, Type N5, Type N6, and Type N8 in the middle of the figure has been set to 60°, 36°, 30°, and 22.5° respectively. The fold of rotational symmetry (n) has been marked in the figure.
Therefore, with the fold of rotational symmetry increasing, the modulation effect of the rotation angle on the $\Delta T$ spectra will disappear gradually. In the wavelength band between 3.3 to 5 $\mu$m, rotation angle cannot change the $\Delta T$ spectra effectively when the fold of rotational symmetry exceeds five.

Selective absorption of circularly polarized waves is realized in this paper, as shown in Fig. 5. The absorption of LCP and RCP waves ($A_{LCP}$ and $A_{RCP}$) (Ouyang, et al. 2018; Cao, et al. 2013; Wang, et al. 2019) and the CD spectra can be calculated as:

\[
A_{LCP} = 1 - R_{RL} - R_{LL} - T_{RL} - T_{LL} = 1 - r_{RL}^2 - r_{LL}^2 - t_{RL}^2 - t_{LL}^2
\]

\[
A_{RCP} = 1 - R_{LR} - R_{RR} - T_{LR} - T_{RR} = 1 - r_{LR}^2 - r_{RR}^2 - t_{LR}^2 - t_{RR}^2
\]

\[
CD = A_{RCP} - A_{LCP}
\]

Here, the $R_{RL}$ ($R_{LR}$) is the cross-polarized reflectance of LCP (RCP) wave, and $R_{LL}$($R_{RR}$) is the co-polarized reflectance of LCP (RCP) wave. The $T_{RL}$ ($T_{LR}$) is the cross-polarized transmittance of LCP (RCP) wave, and $T_{LL}$($T_{RR}$) is the co-polarized transmittance of LCP (RCP) wave.

When the folds of rotational symmetry do not exceed five, it can be found that the rotation angle can still modulate the CD responses. As shown in Fig. 5f, in a period of rotation, the maximum CD response of each type of structure can obtain 0.46, 0.80, 0.56, 0.65, and 0.75 respectively.

In addition, two reconfigure strategies are studied for more flexible designs. For Type N4S1 shown in Fig. 6, the direction of blades in both layers are different from Type N4, when the upper nano-structure is rotated clockwise ($\theta < 0^\circ$), $\Delta T$ spectra are shown in

\[
\frac{\Delta R}{R_0} = \left(1 - \frac{T}{R} \right)\left(1 - \frac{R}{T} \right)\left(1 - \frac{T}{R_0} \right)
\]

\[
\frac{\Delta T}{T_0} = \left(1 - \frac{T}{R} \right)\left(1 - \frac{R}{T} \right)\left(1 - \frac{T}{R_0} \right)
\]

Fig. 5 a–e are the CD spectra of the metamaterial with three, four, five, six, and eight folds of rotational symmetry respectively; (f) are the curves of each type of structures with maximum CD response.
Fig. 6a, which are opposite to Fig. 3e. Similarly, Mode -1, Mode -2, Mode -3, and Mode -4 are used to mark the occasions for rotation angles of 0°, −20°, −42°, and −80° respectively.

By changing the direction of blades in upper layer’s nano-structure of Type N4 only, a unit as Type N4S2 shown in Fig. 6 has been investigated. Metamaterial of this design has significantly different ΔT spectra as shown in Fig. 6b. Following the previous discusses, four different modes, middle and long wavelength bands (Mode 5), short and middle wavelength bands (Mode 6); single wavelength band (Mode 7); short and long wavelength bands (Mode 8), are analyzed in the figure corresponding to the rotation angles of 0°, 10°, 50°, and 80° respectively. It should be noted that different fold of rotational symmetric nanostructures which are discussed in Fig. 4 are able to follow these two reconfigure strategies for more different responses.

To explain the underlying physical mechanism of the proposed metamaterial, the charge distribution of three different modes at middle wavelength band are simulated, as shown in Fig. 7. The direction of the local electric dipoles is marked by the dashed arrow. To simplify the analysis, equivalent electric dipoles (Zhang, et al. 2018; Zhou, et al. 2020) of upper and lower structures have been calculated and marked with two solid arrows below the charge distribution. Based on the Born-Kuhn model (Yin, et al. 2013), when the two layers of equivalent dipoles form an acute angle or are parallel to each other, which means that the antibonding mode is excited; when the equivalent dipoles form an obtuse angle or are antiparallel to each other, which means that the bonding mode is excited.

For Mode 1, when the metamaterial is excited by RCP waves at 4.01 μm, the local electric dipoles in the two parallel nano-rods of lower layer are mainly left pointing and both of the vertical nano-rods of the lower layer have local electric dipoles pointing upward. Similarly, local electric dipoles in the upper layer are pointing right in the parallel nano-rods and pointing upward in the vertical nano-rods. It can be observed that the angle between the two equivalent dipoles is obtuse, so bonding mode is excited. Also, when mode 1 is excited by the LCP wave, it can be found that the equivalent dipole corresponds to the antibonding mode.

To explain why modes switching can be implemented by angle rotation in the proposed metamaterial, the charge distribution at 4.01 μm of Mode 2 and Mode 3 are studied. Under
the excitation of different circularly polarized waves, the local electric dipoles of Mode 2 build two same hybrid modes. Differently, the local electric dipoles of Mode 3 build two different hybrid modes. As a result, the former one almost has no ΔT response, but the latter one has strong ΔT response.

In plasma resonances excited by electromagnetic waves, bonding mode has lower resonance energy than antibonding mode, similar to molecular orbital theory (Li and Zhang 2016; Prodan and Nordlander 2004). More specifically, the phenomenon that bonding modes have higher transmission coefficient than antibonding modes’ as shown in Fig. 3. In a summary, the hybrid mode excited by different circularly polarized waves can be adjusted by changing the rotation angle. The ΔT responses are very weak when the same hybrid modes are excited by different circularly polarized waves. Otherwise, if different hybrid modes excited by different circularly polarized waves, the relatively strong ΔT responses will appear.

It should be noted that when the rotation angle is 0°, the separate two layer of gold structure is no longer 3D chiral. However, each layer of gold structure is placed on a substrate of different thicknesses and different materials, and the gold structure of lower layer is wrapped by polymer film. The whole metamaterial unit (gold structure-polymer-gold structure wrapped by polymer-silica) is still 3D chiral (Arteaga, et al. 2016).

To reveal the advantages of this work more completely, a compassion table is established (Table 2). As for ΔT, it can be seen that in the series of rotational symmetric metamaterials we discussed, different types possess different maximum ΔT and different number of modes can be modulated. Among the different types mentioned in this article, Type N4 has the largest number of modes and can achieve the strongest ΔT response. And through the reconfigure strategies which are proposed in Fig. 6, much more modes of ΔT can be realized. As for CD, it can also be found that strong CD are realized in Type N4 which is relatively high in the similar chiral absorbers (Li, et al. 2014; Wang, et al. 2019).

Due to the limitation of experimental conditions, those structures in this paper haven’t been fabricated. The fabrication process can be realized as follow: firstly, by using electron-beam lithography (EBL), the lower n-fold rotational symmetric structures layer can be fabricated on silicon dioxide substrate. Secondly, polymer is spin-coated...
onto the surface of the first layer and planarized to obtain the second layer. And then, the upper layer of structures can be fabricated by EBL. For more details on sample fabrication, previous researches (Liu, et al. 2008; Yin, et al. 2015) can be referred.

4 Conclusion

In a summary, a series of n-fold rotational symmetric metamaterials are studied in this paper. By changing the rotation angle between the upper and lower nano-structures, various modes of ΔT responses are realized. Meanwhile, reconfigure strategy for opposite ΔT responses, sensitivity of ΔT and CD spectra to rotation angle, and selective absorption are studied in this work. The multi-mode chiroptical responses and the variety of designs and modulation strategies in mid-infrared have proved that this work is beneficial to the applications of multimode circular polarized wave modulation; molecular structure detection; and bio-sensing.

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Declarations

Conflict of interest Not applicable.
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