Original research article

Accurate regulation of circular dichroism signal in double-layer nanostructure

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A B S T R A C T
Chiral plasmonic systems have been shown to exhibit stronger circular dichroism (CD) because of strong interaction between light and noble metals. In this study, we utilize double-layer chiral plasmonic nanostructures composed of achiral nanoplate and two achiral nanodisks to produce a chiroptical response. The chiroptical response is due to the twisted electric dipoles and the phase between them. In addition, the CD signal of the plasmonic nanostructure can be accurately regulated when plasmonic nanostructure parameters are tuned. Overall, results will allow further tuning of the optical responses in plasmonic systems to tailor chiral light–matter interaction.

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1. Introduction

Chiral structures are those that cannot be superposed with their mirror images [1–4]. Chiral metamaterials have revealed several novel electromagnetic phenomena, such as negative refraction [5,6], asymmetric transmission effect [7–10], linear or/and circular polarization conversion [11,12], circular dichroism (CD) effect [13–16] and others. In particular, the CD of circularly polarized light through chiral metamaterials has attracted significant attention in recent years. This effect is due to different transmission intensities between the left circularly polarized light (LCP) and right circularly polarized light (RCP). Chiral structures with CD are always related to circular polarizers for optoelectronic conversion [17,18], polarization conversion [19,20], and molecular analysis [21–24].

In recent years, the CD effect of nanoscale plasmonic systems was widely studied. The nanoscale plasmonic systems have shown significantly enhanced CD because of the strong interaction between light and noble metals [25,26]. These significant interactions between electric and magnetic resonance cause large CD signals in 3D helix or helix-like chiral plasmonic nanostructures [27–32]. Thus, a layer-by-layer chiral nanostructure is designed considering that the helix-like chiral structure is complex and not easy to tune. In addition, the layer-by-layer chiral plasmonic nanostructure can generate larger CD signals because of the coupling between two layers [33–41]. Various approaches have been used to create plasmonic nanostructures and tune chiroptical responses. Changing parameters are often applied to tune the magnitude of a CD signal or shift the wavelength of the aforementioned chiral structures; however, the CD signals cannot be regulated individually. This phenomenon provides a hint to design nanoscale plasmonic systems to accurately regulate optical response without affecting another response.

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In this study, a double-layer chiral nanostructure that consists of an achiral nanoplate and two achiral nanodisks were designed to generate a CD effect. The results show that the double-layer chiral mental nanostructure generates two CD signals, and one CD signal can be tuned by varying the parameter without affecting the other CD signal. This process enables investigating plasmonic analog and studying optical response when the resonances of the constituents are intentionally tuned. In addition, the Born–Kuhn model has been used to analytically interpret the observed CD spectrum, providing insight into the underlying coupling mechanisms. This study can guide the design to accurately tune chiroptical response for practical application in biosensing.

2. Structure and computational method

Fig. 1(a) shows the proposed nanostructure arrays, and each unit is composed of two layers. The bottom layer is two nanodisks and the top layer is a nanoplate. The rotational symmetry centers of the two nanodisks coincide with the center of the nanoplate in the z-direction. The period is \( P_x = 300 \text{ nm} \) and \( P_y = 300 \text{ nm} \) in the x and y direction, respectively. Fig. 1(b) depicts the cell of nanostructure in x-z plane. The radius of nanodisk is labeled as \( R \). The nanoplate has a length of \( L \). The width and height of nanodisks are indicated as \( W = 60 \text{ nm} \) and \( H = 20 \text{ nm} \), respectively. The gap between two layers is \( G \).

Fig. 1(c) shows the cell of nanostructure in y-z plane. The twist angle between the line of center of two nanodisks is \( \alpha \) in the x-y plane. The distance between the centers of the two nanodisks is \( D = 120 \text{ nm} \).

The 3D finite-element method software COMSOL Multiphysics is used to calculate the transmittance of nanostructure arrays. The refractive index of gold is taken from Ref. [42]. The excitation sources are RCP light and LCP light along the \( +z \) direction and the magnitude of the incident electric field is set at 1 V/m. Transmittance was defined as \( T = P_{\text{out}}/P_{\text{in}} \), which is the ratio of output power to incident power. The transmittance spectrum of RCP light and LCP light are represented by \( T_{++} \) and \( T_{--} \), respectively. Therefore, the chirality of structures is represented by CD = \( T_{++} - T_{--} \).

3. Results and discussion

Fig. 2(a) shows the transmission spectra of plasmonic arrays under RCP and LCP light illuminations. The structure parameters of the arrays are \( L = 180 \text{ nm} \), \( R = 30 \text{ nm} \), \( \alpha = 45^\circ \), and \( G = 20 \text{ nm} \). Two evident resonant modes, namely, modes I and II of approximately 860 and 550 nm, respectively, are observed in the transmission spectra. Fig. 2(b) is the CD spectra of the nanostructure arrays. One CD peak and valley appear at the resonance wavelengths. In mode I, the transmittance under LCP illumination is larger than those under RCP illumination. In mode II, the transmittance under RCP illumination is larger than those under LCP illumination. Around the resonance wavelengths, the spectra of \( T_{++} \) and \( T_{--} \) correspondingly present different responses, thereby leading to a large CD effect.

The charge distributions were examined to investigate the transmission mechanism of the nanostructure arrays under circular illumination. Fig. 3 shows the top view of the normalized charge distributions of the nanodisk and nanoplate, where red and blue indicate positive and negative charges, respectively. Fig. 3(a) and (b) shows the normalized charge distributions of the nanoplate at modes I and II, respectively. The green arrows represent the equivalent electric dipole moments of nanoplate (upper layer). Fig. 3(c) and (d) shows the normalized charge distributions of the nanodisks at modes I and II, respectively. The light-blue arrows represent the equivalent electric dipole moments of the nanodisk (lower layer).

At \( \lambda_1 = 860 \text{ nm} \), the equivalent electric dipole moment of the nanodisk and the dipole moment of the nanoplate, along the long axis direction is presented. The effective dipoles \( P_{x-D1} \) and \( P_{x-D2} \) constitute \( P_{x-D} \). In Fig. 3(e), \( P_{y-D} \) and \( P_{y-U} \) form a bonding mode in the Born–Kuhn oscillator model [43,44]. At \( \lambda_1 = 550 \text{ nm} \), the effective dipole \( P_{y-U} \) represents the equivalent electric dipole moment of the nanoplate along the short axis direction. \( P_{x-D1} \) and \( P_{x-D2} \) represent the equivalent electric dipole moments of nanodisks and can also
Fig. 2. (a) Simulated transmittance spectra of nanostructure under circular polarized excitation along $-z$ direction; (b) Simulated CD spectra of nanostructure.

Fig. 3. The charge distributions of the nanostructure for RCP under $-z$ direction light excitation. The charge distributions of nanoplate at (a) mode I and (b) mode II. The charge distributions of nanodisks at (c) mode I and (d) mode II. Born-Kuhn modes of nanostructure at (e) mode I and (f) mode II.
constitute $P_{\text{II-D}}$. In Fig. 3(f), $P_{\text{II-D}}$ and $P_{\text{II-U}}$ form a bonding mode in the Born–Kuhn oscillator model. Most importantly, the resonance mode of the nanodisks occurs at approximately 550 nm and that of the nanoplate is at 860 nm. In addition, we sequentially investigate the influence of varied $\alpha$, $L$, $R$, and $G$ of nanostructure arrays on CD where other parameters are fixed as in the control group. The angle between the two layers was tuned to explore the influence of asymmetry of nanostructure on CD. Fig. 4(a) depicts the simulated CD spectra in different angle $\alpha$ with fixed $L = 180$ nm, $G = 20$ nm, and $R = 30$ nm to investigate the effects of angle $\alpha$ on CD. At $\alpha = 0^\circ$ and $90^\circ$, the structure is achiral, and the two peaks disappear. When $\alpha$ is increased from $0^\circ$ to $150^\circ$, modes I and II do not shift. With an increase in $\alpha$, every equivalent electric dipole moment of components is not changed; thus, modes I and II do not shift.

Fig. 4(b) shows the simulated CD spectra for different length of nanoplate with fixed $G = 20$ nm, $R = 30$ nm, and $\alpha = 45^\circ$ to investigate the effects of length $L$ on chiroptical response. As we can see in Fig. 4(b), mode I red shifted whereas mode II unchanged with the length of nanoplate increased. With an increase in $L$, the equivalent electric dipole moments $P_{\text{II-U}}$ on the nanoplate increased, resulting in red shift of the resonance wavelength. But the equivalent electric dipole moments $P_{\text{II-D}}$ and $P_{\text{II-U}}$ both unchanged, resulting in mode II does not shift. Thus, mode II can be individually tuned by length of the nanoplate.

In Fig. 4(c), the CD spectra is shown for $G = 20$ nm, $\alpha = 45^\circ$, and $L = 180$ nm with different radius of nanodisk. With an increase of $R$, mode II blue shift, but mode I does not shift. In the near field, the component of $P_{\text{I-D}}$ and $P_{\text{II-D}}$ increased with an increase of $R$, thereby resulting in the red shift of mode II. However, the equivalent electric dipole moments $P_{\text{I-U}}$ and $P_{\text{II-U}}$ both unchanged, resulting in mode I does not shift. Thus, mode II can be individually tuned by radius of the nanodisk.

Finally, the distance of two layers was tuned to investigate the influences of the coupling of the chiroptical response. Fig. 4(d) shows the CD spectra of different gap $G$ between two layers with fixed $L = 180$ nm, $R = 30$ nm, and $\alpha = 45^\circ$. At mode I and II, CD spectra undergo a continuous blue-shift for the increasing gap $G$. Given that modes I and II are ascribed to the bonding mode between the electron oscillations on the nanodisk and nanoplate, this finding indicates that attraction exists between nanodisk and nanoplate. An increased $G$ will decrease the attraction between them, leading to a decrease in the equivalent electric dipole moments and resulting in the blue shift of the resonant wavelength. Experimentally, the middle dielectric layer was considered in the design of chiral plasmonic nanostructure. When the middle SiO$_2$ dielectric layer was added, the resonance modes red shifted.

4. Conclusions

A double-layer chiral plasmonic nanosystem with two nanodisks and a nanoplate is proposed to generate a chiral response. The two CD signals can be regulated individually in the nanostructure. Mode I is mainly due to the resonance of the nanoplate along the long axis. Mode II is mainly due to the resonance of the nanodisks. The CD mechanism of the double-layer nanostructure satisfies the Born–Kuhn model for CD effects. Overall, these results are useful for designing a chi-
ral plasmonic nanosystem for a tailored light–matter interaction. This system is expected to be widely applied in biological monitoring, analytical chemistry, and negative refractive-index media.

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