Giant circular dichroism and its reversal in solid and inverse plasmonic gammadion-shaped structures

SHAN WU,1,* PINGPING QU,1 JIANQIANG LIU,2 DANDAN LEI,1 KAIYIN ZHANG,1 SHUTAO ZHAO,1 AND YONGYUAN ZHU3

1Department of Physics, Laboratory of Functional materials and Devices for Informatics, Fuyang Normal College, Fuyang, Anhui 236032, China
2College of Science, Jiujiang University, Jiujiang, Jiangxi 332005, China
3Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, China

*swu@fync.edu.cn

Abstract: Chiral plasmonic structures have been shown to possess large circular dichroism (CD) responses. Here, we investigate the CD responses in a solid and inverse metallic structure composed of a stacked right-twisted gammadion metallic nanoparticle and a left-twisted gammadion nanoaperture array, where a giant circular dichroism is achieved. In addition, the sign of the CD responses can be reversed through the changes of the geometric parameters. Further analysis reveals that the Fabry-Perot (F-P) resonance of cross-polarization conversion of electric field governs the change of the CD. It can be envisioned that our findings will allow further tuning and manipulation of the CD responses for tailored circular polarized light-matter interaction.

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References and links

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1. Introduction

Linearly polarized light can be shown to consist of right and left handed circularly polarized components with equal intensity. For the right (left) handed circularly polarized light (CPL), an observer looking toward the light source will see the electric vector rotating in a clockwise (counterclockwise) sense. Circularly polarized light is chiral, that is, cannot be superimposed onto its mirror image. A chiral molecule has different absorption when illuminated with the (counterclockwise) sense. Circularly polarized light is chiral, that is, cannot be superimposed onto its mirror image. A chiral molecule has different absorption when illuminated with the (counterclockwise) sense. Circularly polarized light is chiral, that is, cannot be superimposed onto its mirror image.

A chiral molecule has different absorption when illuminated with the left or right CPL, this effect called circular dichroism (CD) [1, 2]. CD spectrum reveals chiral behavior in optical transmission of sandwich coaxial square ring arrays,” Appl. Phys. Lett. 93(10), 101113 (2008).


arrangements of metallic nanoparticles [12,13]. The conventional chiral structures are built up by the metallic nanoparticles, such as rod particle [14], L-shaped particle [15], gammadion-shaped particle [16–18], and the others [19, 20], on which the localized plasmonic resonance can be excited. The interaction of the plasmonic resonances in individual nanoparticle or its component via the strongly localized electric fields enhances the CD response [21, 22]. On the other hand, based on Babinet's principle, an inverse metallic structure, i.e. a nanoaperture array in a continuous metallic film also exhibits cavity plasmonic resonance with the strongly localized electric field. In complex plasmonic nanostructures consisted of solid and inverse metallic nanostructure, elemental plasmon modes can also couple and exhibit novel properties, such as electromagnetically induced transparency (EIT) [23], dispersionless optical activity [24, 25], asymmetric transmission and optical rotation [26]. In this work, we investigate the CD response of solid and inverse gammadion-shaped metallic structure. A giant dissymmetry factor \( g = 1.58 \) can be obtained at wavelength 922 nm, which is three orders of magnitude higher than those found in common biomolecules. Furthermore, we also show that the sign of the CD spectrum may be controlled by tuning the relative distance between the solid and inverse gammadion-shaped structures. Further analysis reveals that the Fabry-Perot (F-P) resonance of cross-polarization conversion of electric field governs this change.

2. Material design and calculation

The three-dimensional schematic view of the solid and inverse gammadion-shaped metallic structure is shown in Fig. 1(a), with the geometrical parameters illustrated in Fig. 1(b). The whole system consists of two metallic structures separated by the silica with thickness \( d \): one being a right-twisted gammadion metallic nanoparticle array embedded in silica substrate and the other being a left-twisted gammadion nanoaperture array in metallic film covering the sample surface. A circularly polarized light propagates in a positive \( z \) direction. The CD spectra are calculated by the finite-difference time-domain (FDTD) method [27]. The Drude-model dielectric function is used for metal (silver) with a plasma frequency of \( 1.374 \times 10^{16} \) rad/s and a collision frequency of \( 2.02 \times 10^{14} \) rad/s [28]. The permittivity of silica substrate is set as 2.25, and background environment is assumed to be air with a permittivity of unity.

![Fig. 1. (a) Three-dimensional schematic view of the solid and inverse gammadion-shaped metallic structure. (b) Geometrical parameters of the unitcell: period \( P = 700 \) nm, \( l = 300 \) nm, \( w = 100 \) nm and the thickness of gammadion-shaped metallic particle and aperture \( t = 50 \) nm with the distance between the upper and lower layers \( d = 160 \) nm.](image-url)
3. Results and discussion

In our calculation, the CD is defined as

\[ \text{CD} = A_r - A_l = T_{ll} - T_{rr} \]  

(1)

where \( A \) and \( T \) is the absorbance and transmittance for the right \((rr)\) and left \((ll)\) CPL incident waves (the first and the second subscript means out and in, respectively.), which are given by \( A = 1 - R - T \), where \( R \) is reflectance. The equality in (1) follows from \( R_{rr} = R_{ll}, R_{rl} = R_{lr} = 0 \), and \( T_{rl} = T_{lr} = 0 \), which can be guaranteed from the reciprocity theorem and the \( C_4 \) rotational symmetry of the structure, respectively [16, 17, 29]. Figure 2(a) shows the transmittances of right \((T_{rr})\) and left \((T_{ll})\) CPL and the CD spectrum. There are two CD peaks at wavelengths about 778 nm and 922 nm. For these two peaks, their amplitudes reaches 0.32 and 0.51, and the dissymmetry factors \( g \) are 1.17 and 1.58, respectively, which are larger than those of single layer and bilayer solid gammadion structures [18].

![Figure 2](image)

Fig. 2. (a) The calculated right (black solid line) and left (red dash line) circular polarized transmission spectra and the CD spectrum (blue short dash dot line) of the system. (b) The calculated electric field \( E_{xy} \) (black solid line), \( E_{yy} \) (red dash line) components and the phase difference (blue short dash dot line) of the transmitted light with the y-polarization incident light.

To investigate the physical mechanism of the above results, we convert the circular polarized transmission into linear polarized transmission. Based on the Jones matrices [30] we have

\[
\begin{pmatrix}
    t_{rr} & t_{rl} \\
    t_{lr} & t_{ll}
\end{pmatrix} = \frac{1}{2} \begin{pmatrix}
    (t_{xx} + t_{yy}) + i(t_{xy} - t_{yx}) & (t_{xx} - t_{yy}) - i(t_{xy} + t_{yx}) \\
    (t_{xx} - t_{yy}) + i(t_{xy} + t_{yx}) & (t_{xx} + t_{yy}) - i(t_{xy} - t_{yx})
\end{pmatrix}
\]  

(2)

where \( t_j \) is the transmitted coefficient, and the subscript \( ij \) stands for incident polarization \( j \) and transmitted polarization \( i \). For a Lorentz-reciprocal structure, the scattering matrix must
be symmetric, requiring \( t_{xy} = -t_{yx} \) \([21, 29–31]\). Furthermore, our structure has the \( C_4 \) rotational symmetry with \( t_{xx} = t_{yy} \) \([29]\). And thus, \( t_{xy} = t_{yx} = 0 \) in the Eq. (2). The CD can be calculated as

\[
\text{CD} = |t_x|^2 - |t_y|^2 = -2 \text{Im}[t_x^*(t_{xy} + t_{yx})] = -4E_{yy}E_{xy} \sin \Delta \phi \tag{3}
\]

where \( E_{yy} \) and \( E_{xy} \) are the normalized electric field amplitudes in the linear polarization transmission, and \( \Delta \phi = \phi_{yy} - \phi_{xy} \) is the phase difference. From Eq. (3), it is obvious that the prerequisite for CD is non-zero \( E_{yy} \), \( E_{xy} \) and \( \Delta \phi \). Generally, for the conventional single-layer metallic perforated structures, the excited surface plasmon polaritons (SPPs) cannot produce the linear polarization transmission conversion, i.e. \( E_{yy} = 0 \). It is because the incident linear-polarized light cannot excite the SPPs modes along the direction perpendicular to the incident electric field. However, for the special structure, the excited localized surface plasmons (LSPs) can drive the induced charges into another direction, forming linear polarization conversion, i.e. \( E_{yy} \neq 0 \) \([28]\). Here, in our proposed structure, utilizing the scattering of the LSPs excited in lower gammadion-shaped metallic particles, the excited SPPs modes in upper metallic film is not only along the direction of the incident electric field but also along the direction perpendicular to the incident electric field. Figure 2(b) shows the normalized \( E_{yy} \), \( E_{xy} \) and \( \Delta \phi \) spectra in far field. It can be seen from Fig. 2(b) that the spectral line shape is similar to that of the enhanced optical transmission (EOT) in metallic perforated structures \([32]\). There are three peaks \( A \) (776 nm), \( B \) (943 nm) and \( C \) (1083 nm) in \( E_{xy} \) spectrum corresponding to the SPPs \( A(1,0), G(1,1) \) and \( G(1,0) \), where \( A \) stands for air-metal surface and \( G \) for silica-metal surface, \((m,n)\) is the reciprocal vector.] For the \( E_{yy} \) spectra, the peaks \( A \) and \( B \) exist, however, peak \( C \) disappears. The phase difference \( \Delta \phi \in (0^\circ, 180^\circ) \) in the wavelength range from 750 nm to 1058 nm. Thus, based on Eq. (3) we can get a large positive CD effect. However, for the peak \( C \) there is a near-complete cross-polarization conversion, i.e. \( E_{yy}\approx0 \), resulting in a near-zero CD. It is due to the destructive interference of electromagnetic wave emitted from the different zone on the upper surface. Figure 3 represents the electric field \( E_{xy} \) and \( E_{yy} \) components of the peaks \( A, B \) and \( C \) on the air-metal surface. Unlike the electric field \( E_{xy} \) and \( E_{yy} \) components of peaks \( A \) and \( B \), the \( E_{yy} \) component of peak \( C \) have an opposite phase in the zone (1) and zone (2) \([Fig. 3(f)]\). These act as two anti-parallel dipoles whose emitted electromagnetic waves interfere destructively in far field, leading to a near-zero \( E_{yy} \) component.

In addition, we discover that the sign of the CD spectrum may be controlled by tuning the distance between the two metallic layers. Figure 4(b) depicts the two-dimensional grayscale image of the CD with distance \( d \). There is an inversion of the CD sign when the distance \( d \) increases. In order to survey this change of the CD, we plot the CD spectrum at wavelength 923 nm \([Fig. 4(c)]\). As the distance \( d \) increases, the CD value firstly increases to 0.52 at \( d = 160 \) nm, and then decreases, even vanishes completely at \( d = 260 \) nm. When \( d \) is larger than 260 nm, the CD sign is inverted. From Eq. (3), one can conclude that the sign of the CD is determined by the phase difference \( \Delta \phi \). Thus, we show the CD spectra and the phase difference \( \Delta \phi \) for the thickness \( d = 160 \) nm and 340 nm in Fig. 4(a). Different from the sample of \( d = 160 \) nm, the phase difference \( \Delta \phi \in (0^\circ, 180^\circ) \) in the wavelength range from 750 nm to 1058 nm as \( d = 340 \) nm. It causes the inversion of the CD sign.
Fig. 3. The electric field $E_{xy}$ (a-c) and $E_{yy}$ (d-f) components of peaks $A$, $B$ and $C$ on the air-metal surface, respectively.

The change of the CD sign represents the different interaction between the right- and left circularly polarized light and metallic structures. It is one of important physical features of our mixed structures. The conventional reversals of the CD sign have three cases. Firstly, when a chiral structure changes its handedness, its CD sign can be reversed without the deep physical reasons [18]. Secondly, the reversal of the CD sign occurs in different wavelength range due to the different coupling mode between the localized electric and magnetic resonances [21]. Thirty, the sign of the CD at a given wavelength can be controlled by tuning the relative wavelength of the plasmonic hybridized mode [15] or the conductivity of dielectric in hybrid metamaterial [33]. To reveal the underlying mechanism for the change of the CD sign in our proposed structures, we plot the two-dimensional greyscale image of the electric field component $E_{xy}$ with the distance $d$ in Fig. 5. A dark line with $E_{xy} = 0$ in Fig. 5 (red circular line) is just the dividing line between the negative and positive CD in Fig. 4(b), which depends linearly on the distance $d$, which is a Fabry-Perot (F-P) resonance mode with the effective index $n_{eff} = 1.68$. 
Assumed that the system is illuminated by the $y$-polarized plane wave propagating in the positive $z$ direction. The electric field $E_{\text{Sp}}$ is reradiated by the SPPs propagating along the $x$ direction (defined as SPP$_x$) in the upper metallic film excited by the scattering component $E_{\text{LSP}_x}$ of the LSPs resonances produced in lower gammadion-shaped metallic particles. $E_{\text{LSP}_x}$ in the positive $z$ direction can be written as

$$E_{\text{LSP}_x} = E \cos(\omega t + \frac{2\pi n_{\text{eff}}}{\lambda} z)$$

(4)

Via the reflection of the upper metallic film, it would be given by

$$E_{\text{LSP}_x} = E \cos(\omega t - \frac{2\pi n_{\text{eff}}}{\lambda} z + \pi)$$

(5)

where $n_{\text{eff}}$ is the effective index. Thus, total electric field would be given by

$$E_{\text{LSP}_x} = E_{\text{LSP}_x} + E_{\text{LSP}_x} = 2E \sin\left(\frac{2\pi n_{\text{eff}}}{\lambda} z\right) \cos\left(\frac{\omega t + \pi}{2}\right)$$

(6)
When the distance between the two metallic layers \( d = z_0 = \lambda/2n_{\text{eff}} \), the stationary wave (i.e. Fabry-Perot resonance) is formed with \( E_{\text{LSP}} = 0 \) in the upper metallic film. Note that no SPPs is excited at this condition. Thus, the \( E_{xy} = 0 \), and CD = 0.

\[
\text{As } d = z_0 + z',
\]

\[
E_{\text{LSP}} = 2E \sin \left( \frac{2\pi n_{\text{eff}} z'}{\lambda} \right) \cos \left( \alpha \pi + \frac{\pi}{2} + \pi \right) \]

(7)

Compared to \( d < z_0 \) [Eq. (6)], the phase increases a \( \pi \) radian. Subsequently, the phase difference \( \Delta \phi \) decreases a \( \pi \) radian, resulting in an inversion of the CD sign. Figure 5 also shows a clear anticrossing behavior with the splitting energy of 40 meV, which could be attributed to a strong coupling [34, 35] between the F-P modes and the Rayleigh diffraction modes [36–38] in the metal-silica interface.

Fig. 5. The two-dimensional greyscale image of the electric field component \( E_{xy} \) with the displacement \( d \) of the middle silica layer. The red circular and green square lines represent the F-P resonance and Rayleigh diffraction modes, respectively.

4. Conclusion

In conclusion, we have proposed a solid and inverse metallic structure composed of a right-twisted gammadion metallic nanoparticle array and a left-twisted gammadion nanoaperture array. A strong circular dichroism can be obtained. And the sign of the CD responses can be reversed through the increases of the distance between the two metallic layers. Further, we have also discussed the physical mechanism of circular dichroism, and demonstrated that the changes of the CD are heavily dependent on the polarization conversion \( E_{xy} \). Our work provides a fundamental understanding of circular dichroism in the complex plasmonic structures as well as the potential applications in switchable metamaterials.

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