Helical dichroism for hybridized quadrupole plasmon modes in twisted metal nanorods

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Abstract

Helical dichroism (HD), based on the interaction between chiral plasmonic nanostructures and light with orbital angular momentum (OAM), has attracted researchers in a wide range of fields from the viewpoint of fundamental physics and applications. However, the relation between the HD and the excited plasmon modes has been poorly understood in experiments. Because of the weak chiral interaction between the chiral structures and OAM light, the structure size had to be much larger than the incident light wavelength to obtain a sufficient HD signal in an experiment, resulting in a complex superposition of higher-order plasmon modes. Recently, we experimentally demonstrated that a twisted gold nanorod dimer, one of the simplest 3D chiral plasmonic structures, exhibits giant circular dichroism due to strong plasmon coupling between the nanorods, followed by the hybridization of dipole mode. In this study, we reveal that the HD of this nanorod dimer appears due to the hybridization of quadrupole plasmon mode rather than dipole mode. Furthermore, the measurement of the HD signal can be achieved by using the array of the twisted dimers. The dependence of the HD on the incident light wavelength exhibits that the HD sign changes around the quadrupole plasmon resonance, which is in good agreement with the simulation. These results open new avenues for the profound understanding of the light-matter interaction with respect to angular momentum.

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1 Introduction

The interaction between chiral matter and light has attracted researchers from a wide range of fields, both in fundamental physics and in applications. In particular, the chiroptical response, which depends on the handedness of left- and right-circularly polarized light, has been crucial in biology, chemistry, and pharmaceuticals\cite{1}\cite{2}. However, its signal is generally weak for chiral molecules because of the size mismatch with the light wavelength. With the recent development of fabrication technology, the chiroptical response of artificial nanostructures has been studied\cite{3}\cite{4}\cite{5}. The size of the nanostructure is close to wavelengths of operating visible light, and its design mimics the essence of molecular features, such as chirality and polarization. Typical examples are nanophotonic structures\cite{6}\cite{7}, plasmonic nanoparticles\cite{8}, and chiral metamaterials\cite{9}\cite{10}\cite{11}\cite{12}, which have significantly accelerated the understanding of the chiroptical response.

Light has an angular momentum, which can be considered similar to chirality in matter. As circularly polarized light has spin angular momentum(SAM), optical vortex beam such as Laguerre-Gaussian beam has orbital angular momentum(OAM) \cite{13}\cite{14}\cite{15}\cite{16}. The SAM and OAM are defined by different physical parameters: polarization and spatial phase distribution, respectively\cite{17}. Therefore, OAM has features that would be more strongly dependent on the spatial profile of light. With respect to degrees of freedom, there are two types of SAM, corresponding to left- and right-circular polarization. On the other hand, OAM has infinite degrees of freedom $l = 0, \pm 1, \pm 2, \cdots$, where $l$ stands for the topological charge, which represents the number of helical wavefronts in each wavelength apart. One of the interesting properties of OAM light is that its helical plane of the electric field allows multipole excitation forbidden in dipole approximation. Indeed, this multipole excitation by the OAM light has been demonstrated in some platforms: trapped ions \cite{18}, and two-dimensional metallic structures \cite{19}\cite{20}. In particular, the electric quadrupole excitations could engage the OAM light with chiral matter \cite{21}.

The interaction between the chiral matter and the SAM light has been widely studied in terms of circular dichroism (CD)\cite{1}\cite{2}, which is defined as the difference in absorption for different handedness of circular polarization. Helical dichroism (HD) is an expansion of the CD and refers to the difference in absorption that depends not only on the SAM but also on the OAM\cite{22}\cite{23}. Exploring the HD could deepen the understanding of how chiral matters interact with the angular momentum of light. In recent years, experimental observations of the HD have been realized in various chiral systems, such as the complex mixture of chiral molecules and plasmonic substrates \cite{24}, microstructures with phase distributions of dimensions close to those of the incident vortex beam\cite{25}\cite{26}, the 2D microstructure\cite{27}, and disordered molecular media by using hard X-rays\cite{28}.

Despite this recent progress, an understanding of the experimental HD as regards the mode in the plasmonic structures or molecules is still insufficient. In some previous studies, the analysis of the excited plasmon modes is difficult due to the superposition of the multipolar contributions included in the structure larger than the light wavelength. To approach the molecular situation, the plasmonic structure should be smaller than the operating light wavelength, which results in simple interaction between the chiral structure and light. In
a nanohelix structure, which is smaller than light wavelength, dipolar HD that
events the same spectral behavior as CDs has been observed because the in-
cident OAM is partially converted to the SAM by light-focusing. [25]. Although
Brullot et al. [24] discussed a relationship between HD and electric quadrupole
modes, the structure, the complex mixture of chiral molecules and plasmonic
substrates, is too complex to analyze the relationship.

A twisted nanorod dimer is one of the simplest 3D chiral plasmonic struc-
tures. Recently, we demonstrated that the twisted nanorod dimer with a smaller
size than the light wavelength exhibits the giant chiroptical response of SAM
light for its hybridized dipole plasmon modes[29]. Furthermore, R.M. Kerber et
al. numerically studied the HD for the hybridized dipole modes and showed that
a single twisted nanorod dimer has no HD signal and that the HD signal appears
in the interactions between multiple dimers [30]. In this Letter, we numerically
and experimentally explore the HD for hybridized quadrupole plasmon modes
in the twisted nanorod dimer. Unlike the dipolar HD, the quadrupole HD signal
can be observed even in a single nanorod dimer. Furthermore, the quadrupole
HD signal is enhanced when the dimers are arranged in a periodic array, and
its signal is more pronounced than the dipolar HD signal that appeared in the
dimer array. In addition, we measure the wavelength dependence of the HD and
CD signals of the dimer array and experimentally demonstrate that the sign of
the HD signal changes near the quadrupole resonance but not for the CD signal,
in good agreement with the numerical results.

2 Results and Discussion

We start by analyzing the coupling of light with SAM and OAM to the plasmonic
nanostructure, respectively. In this research, we adopt a golden twisted nanorod
dimer as a chiral plasmonic nanostructure. Figure 1 shows a twisted nanorod
dimer used in the simulation and experiment: $L = 300$ nm, $W = 60$ nm,
$H = 40$ nm, $\theta = 30^\circ$, and $d = 100$ nm. We define the absorption cross-section as

$$A_{abs} = \int_V QdV,$$

where $Q$ is the power loss density in the twisted nanorod dimer and the integral
is taken over their volume $V$. To compare with achiral structures, we also
performed the simulation of nanorod monomers in the following sections.

Firstly, we performed the simulation with circularly polarized light. We
applied left- and right-circularly polarized light (spin angular momentum is
$s = \pm 1$). Figure 1 shows the absorption cross-section $A_{abs}(\lambda)$ of a nanorod
monomer and a twisted nanorod dimer as a function of incident wavelength at
different $s = \pm 1$. In both left- and right-circular polarizations ($s = \pm 1$), there
are plasmon resonance peaks in the spectrum at $\lambda = 1592$ nm. In the inter-
action of circularly polarized light with a sub-wavelength-sized chiral structure,
a chiral dipole is excited as a consequence of the chiral geometry of the illu-
minated structure. The peak in the absorption spectrum of nanorod monomer
corresponds to the plasmonic dipole mode (Figure 1). In the case of the twisted
nanorod dimer, the close presence of nanorods with excited dipole modes in the
peak region causes hybridization, resulting in the observation of a large CD, which is the difference in absorption with different handedness $s = \pm 1$ [30] [29].
Around \( \lambda = 850 \text{ nm} \), there are no peaks with \( s = \pm 1 \). In terms of (CD), there is a certain CD, but its sign does not change around \( \lambda = 850 \text{ nm} \).

Next, we performed the simulation with the light carrying OAM. We applied incident light consisting of linearly polarized (spin angular momentum is \( s = 0 \)) Laguerre-Gaussian mode, whose topological charge is \( l = 1, -1 \), and a beam radius is 2 \( \mu \text{m} \) (Figure 1b). Figure 1b displays the absorption spectrum \( A_{abs}(\lambda) \) with no spin angular momentum (\( s = 0 \)) and the different signs of topological charges. In the case of a nanorod monomer, there is a peak around \( \lambda = 845 \text{ nm} \) in both topological charge \( l = \pm 1 \), which can correspond to the plasmonic quadrupole mode based on the phase distribution (not shown here). In the case of the twisted nanorod dimer, when two nanorods are twisted and close, the dimer with excited quadrupole modes in the peak region hybridizes and forms a bonding mode (in-phased) and anti-bonding (out-phased) mode as in the case of dipole modes (Figure 1f,g). The bonding mode and anti-bonding mode can couple to the light with \( l = \pm 1 \) selectively, leading to the different absorption spectrum: a single resonance peak around \( \lambda = 870 \text{ nm} \) (\( l = +1 \)) and two resonance peaks around \( \lambda = 830, 870 \text{ nm} \) (\( l = -1 \)). The resonance peak of the bonding (anti-bonding) mode of the twisted nanorod dimer is shifted from the quadrupole peak of the nanorod monomer to red (blue) in the absorption spectrum. Therefore, the difference in absorption with different topological charges \( l = \pm 1 \), which is defined as HD, shows a sign change at about \( \lambda = 840 \text{ nm} \). This trend is entirely different from that of CD, which does not show a sign change of CD around \( \lambda = 840 \text{ nm} \). Moreover, these results agree with previous research, which argues that multipole modes can cause HD [26]. There, the structure was too complicated to discuss this relationship clearly. In contrast, our simple plasmonic structure has a quadrupole mode, which is the simplest multipole mode. Then the simulation results show that quadrupole mode and its hybridization induce HD signal.

Furthermore, we also studied the two-dimensional array of those twisted nanorod dimers. Figure 2a shows the configuration of the array in the simulation. Each twisted nanorod dimer in the array has the same configuration as the single twisted nanorod dimer in the previous paragraph (Figure 1a). The distance between these components is 500 nm (Figure 2a). The black dashed line in Figure 2a illustrates the beam width, and the array of structures extends over an area larger than the beam width.

Figure 2b displays the absorption spectrum of the array of twisted nanorod dimers for linearly polarized Laguerre-Gaussian beam along the y-axis (\( s = 0 \), \( l = \pm 1 \)). The spectrum has four peaks, two peaks around \( \lambda = 850 \text{ nm} \) and two peaks around \( \lambda = 1500 \text{ nm} \), in both topological charges \( l = \pm 1 \) (Figure 2b inset). Based on the comparison with the results of a single nanorod dimer (Figure 1c,e), even with the arrayed structure, we observed the hybridization of dipole and quadrupole plasmon modes, respectively. Regarding dipole plasmon modes and their hybridization, in the case of the array, the dipole modes are excited by the effect of dimers far from the beam center, unlike in Figure 1c. The electric field of the Laguerre-Gaussian beam far from its center has a more gentle phase gradient than that at the center. This is more like a plane wave with linear polarization, resulting in the dipole-like plasmonic response of the twisted nanorod dimers than that at the center. Incident light with different values of \( l = \pm 1 \) has the same SAM (\( s = 0 \)), so no strong selective coupling occurs for the hybridized and split dipole modes as in Figure 1a. On the other hand,
Figure 1: The simulation results in twisted nanorod dimer and nanorod monomer and their CD/HD signal (a) The configuration of the twisted nanorod dimer (b) The concept of helical dichroism (HD). The HD is defined as the difference of the absorption with left and right helicities ($l = \pm 1$) (c) The normalized absorption spectrum of the simulation result with circularly polarized light ($s = \pm 1, l = 0$). The colored solid lines show the result of the twisted nanorod dimer, and the dashed line shows that of the nanorod monomer, which amplitude is twice multiplied for comparison. (d) The phase distribution with circularly polarized light. At $\lambda = 1592$ nm, it indicates a dipole-like response. (e) The normalized absorption spectrum of the simulation result with topological charge ($s = 0, l = \pm 1$). The colored solid lines show the result of the twisted nanorod dimer, and the dashed line shows that of the nanorod monomer, which amplitude is twice multiplied for comparison. (f)(g) The phase distribution at two different wavelengths and topological charges are shown. At $\lambda = 870$ nm and $l = +1$, it indicates an in-phased quadrupole response, whereas, at $\lambda = 830$ nm and $l = -1$, it displays an out-phased quadrupole response.

regarding quadrupole plasmon modes and their hybridization, the effect of the dimer near the beam center causes absorption peaks around $\lambda = 850$ nm similar to the peaks in Figure 1. It should be noted that compared to the case of the single twisted nanorod dimer, the absorption cross-section is 50 times larger in the case of the array, such that experimental detection could be realized.

Figure 2 shows the HD spectrum. Around $\lambda = 850$ nm, there is a sign change in HD due to the hybridization of plasmonic quadrupole modes and their selective coupling to the light with $l = \pm 1$. Although the absorption of dipole-like responses is much larger than that of a quadrupole, its HD is much smaller than that of the quadrupole. Conversely, suppose a sign change is observed in HD experimentally in this wavelength range. In that case, it is evidence that the hybridization of plasmonic quadrupole modes and their selective coupling to the light with $l = \pm 1$ is occurring.
Figure 2: The simulation results with the array of twisted nanorod dimers (a) The simulation setup with 21 twisted nanorod dimer. The dashed line circle displays the beam width of 2 µm. (b) The normalized absorption spectrum of the array of twisted nanorod dimer (s = 0, l = ±1). The inset plot focuses on the wavelength around λ = 850 nm. (c) The HD signal spectrum of Figure 2b. The inset plot focuses on the wavelength around λ = 850 nm.

To experimentally demonstrate the sign change in HD related to the hybridization of plasmonic quadrupole modes and their selective coupling to the light with l = ±1, we measured the wavelength dependence of HD for a plasmonic nanostructure. Firstly, we tried HD measurement using a single twisted nanorod dimer, but it failed due to the small signal-to-noise ratio. Based on the discussion about the simulation using the array structure in the previous paragraph, the array of twisted nanorod dimers was used instead of a single twisted nanorod dimer to amplify the signal-to-noise ratio to a detectable level in the experiments. The array has the same geometry as in the simulation of the array of twisted nanorod dimer (Figure 3a). It is a two-dimensional lattice of 500 nm apart twisted nanorods. The experimental setup is shown in Figure 3b. The spatial light modulator (SLM) generated light with OAM as a Laguerre-Gaussian beam, and the light was incident into the sample on the piezo stage. The SLM is controlled through the computer, and the topological charge is switched electrically. The polarization of light can be controlled through the half- and quarter-wave plates. In addition, the piezo stage scanned the sample in a two-dimensional direction with a resolution of 100 nm. To confirm the generation of the desired vortexed wavefronts, we did the imaging of their wavefronts with the interferometer (See Supplementary Information).

Two types of light were incident on the sample. One is circularly polarized light without topological charge (s = ±1, l = 0). The other is linearly polarized light along the bottom nanorod (longitudinal in Figure 3b) with topological charge (s = 0, l = +1, −1). To compare the optical responses of chiral and achiral structures, we also measured the array of nanorod monomers. The absorption signal A(r, λ) is defined as:
\[ A(r, \lambda) = 100 \times \frac{T_{\text{ref}} - T_{\text{sample}}(r, \lambda)}{T_{\text{ref}}} \]  \hspace{1cm} (2)

where \( T_{\text{sample}} \) and \( T_{\text{ref}} \) are the transmission intensity with and without the nanorod region, respectively. The CD signal, \( S_{\text{CD}} \), and the HD signal, \( S_{\text{HD}} \), are defined as:

\[ S_{\text{CD}}(r, \lambda) = A_{s=-1}(r, \lambda) - A_{s=+1}(r, \lambda) \]  \hspace{1cm} (3)
\[ S_{\text{HD}}(r, \lambda) = A_{l=+1}(r, \lambda) - A_{l=-1}(r, \lambda). \]  \hspace{1cm} (4)

Figure 3c displays the 2D plot of \( S_{\text{CD}}(r, \lambda) \). The plots appear uniform with no sign change. The average \( S_{\text{CD}}(\lambda) \) over 2D plot region is plotted in Figure 3d. While the array of twisted nanorod dimers shows some CD, that of the nanorod monomer is negligibly small. Figure 3d shows the 2D plot of \( S_{\text{HD}}(r, \lambda) \). Unlike the 2D plot of \( S_{\text{CD}}(r, \lambda) \) (Figure 3c), the \( S_{\text{HD}}(\lambda) \) of the array of twisted nanorod dimer indicates a certain sign change around \( \lambda = 820 \) nm. On the other hand, the \( S_{\text{HD}}(\lambda) \) of the array of nanorod monomer is relatively smaller and looks more constant than that of twisted nanorod dimer (Figure 3d). This spectral behavior is in good agreement with the simulation results of the array of twisted nanorod dimers, especially with respect to the sign change of HD (Figure 2b, 3f).
Figure 3: The experiment setup and results. (a) The SEM image of the sample. The dashed white line rectangular displays a 2D plot region (2 µm × 2 µm). (b) The experiment setup. The light from Ti-Sapphire laser passes through a half wave plate (HWP) and linear polarizer (LP) to set an appropriate polarization before the spatial light modulator (SLM). The SLM generates light with OAM as a Laguerre-Gaussian beam, and it is introduced onto the sample on the piezo stage. The transmission light is injected into the photodetector via optical fiber. Sub-images show the intensity profile of the Laguerre-Gaussian beam ($l = \pm 1$) before the objective lens. (c) The 2D plot of the CD signal at five different wavelengths with twisted nanorods and mono nanorod. (d) The spectrum of the average CD signal over 2D plot regions. (e) The 2D plot of the HD signal at five different wavelengths with twisted nanorods and mono nanorod. (f) The spectrum of the average HD signal over 2D plot regions.

In summary, the present work reports two achievements: the demonstration of the fact that the twisted nanorod dimer and its array, even whose size is smaller than the wavelength of the light, have the HD signal with the simple excitation mode, the experimental verification of the HD signal with the array of twisted nanorod dimer. The present work also opens a new window into HD studies. Such studies have previously only been achieved by the single 2D plasmonic structure, which size is larger than the wavelength of the incident light and the assembly of molecules where the excitation mode is ambiguous. The present work demonstrates that even the plasmonic structure, which is smaller than the wavelength of the incident light, has a certain HD, and its excitation mode is quite simple. Furthermore, considering the array of these structures, the signal-to-noise ratio can be amplified to a level where experimental detection is possible. These achievements promise exciting developments in the fundamentals of the interaction between chiral matter and light with OAM, which pave the way to novel insights into the profound understanding of the light-matter interaction with respect to the angular momentum.
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