A chiral near-field with a highly contorted electromagnetic field builds a bridge to match chiral molecules and light wavelengths with large size differences. It significantly enhances the circular dichroism of chiral molecules and has great prospects in chirality sensing, detection, trapping, and other chirality-related applications. Surface plasmons feature outstanding light-trapping and electromagnetic-field-concentrating abilities. Plasmonic chiral nanostructures facilitate light manipulation to generate superchiral near-fields. Meanwhile, the nanophotonic structures have attracted significant interest to obtain strong chiral near-fields due to their unique electromagnetic resonant properties. During the interaction of light and chiral materials, the chiral near-field not only bridges the light and chiral molecules but is also responsible for the optical activities. This paper reviews state-of-the-art studies on generating or enhancing chiral near-fields using plasmonic and photonic nanostructures. The principle of chiral near-fields and the development of chiral near-fields with plasmonic and photonic nanostructures are reviewed. The properties and applications of enhanced chiral near-fields for chiral molecule detection, spin-orbit angular interaction, and the generation of the chiral optical force are examined. Finally, current challenges are discussed and a brief outlook of this field is provided.

1. Introduction

Chirality refers to the symmetrical nature of objects. A chiral object has a 3D structure that cannot be superimposed on its mirror image by rotation or translation, that is, human hands. Objects with opposite chirality are called enantiomers. They are identical in chemical composition and have the same scalar physical properties, that is, density, enthalpy of formation, and vibrational frequency. Their differences are only observable when they interact with other chiral objects or fields. Chiral objects are ubiquitous in nature, ranging from sub-atomic particles to galaxies. Chirality is also essential to human lives because most biomolecules, the building blocks of life, including amino acids, nucleic acids, and carbohydrates, exist only in one chiral configuration.

The absorption difference, that is, circular dichroism (CD), is widely used to distinguish the chiral states of molecules based on the different interactions of molecular enantiomers with left-hand and right-hand circularly polarized light (CPL), which is a chiral electromagnetic field. However, the CD of small chiral molecules is very weak because the absorption cross sections of left and right CPL differ by less than one in a thousand. This is a result of the size mismatch of the molecules and the light wavelength. At the scale of small molecules, the CPL field exhibits only a small change in the helical pitch with a slight twist, resulting in weak excitation perturbation.

Two strategies have been used to enhance the CD intensity. The first focuses on the molecular aspect of CD and designs molecules with a large optical dissymmetry. The second uses exciting electromagnetic fields that are more contorted than commonly used CPL fields. Comparing with the first strategy, the second one has superior versatility without the limitation of measured molecules. Following this strategy, Tang and Cohen proposed to enhance the field dissymmetry by shortening the field line reorientation distance relative to the free-space wavelength. They were the first authors to propose the chiral near-field in 2010 to obtain “superchiral light” with a highly distorted electromagnetic field. They obtained a light field with chiral asymmetry much greater than that found in CPL plane waves at the nodes of an optical standing wave. Excited by the highly distorted “superchiral light,” they conducted an experiment and observed an 11-fold enhancement over CPL in the discrimination of the enantiomers of a biperylene derivative.
Localized surface plasmons (LSPs) exhibit outstanding light-trapping and electromagnetic-field-concentrating abilities.[8] Therefore, chiral plasmonic nanostructures were proposed as an effective and reliable approach to obtain a chiral near-field in the vicinity of the nanostructures.[9] The principles for designing chiral plasmonic nanostructures with strong chiral near-fields were proposed.[10] In recent years, different types of plasmonic nanostructures have been designed to generate chiral near-fields, ranging from planar to 3D, monomer to polymer, homogeneous to heterogeneous, and chiral to achiral structures. The excitation light was also expanded from CPL to linearly polarized light.[11–16] Similarly, some nanophotonic structures have been proposed to produce chiral near-fields due to their strong electromagnetic responses and low loss.[17–21] Chiral near-fields have been proposed to produce chiral near-fields due to their strong helicity of any divergenceless vector field \( \mathbf{B}(r) \) in a domain \( \mathcal{D} \subset \mathbb{R}^3 \) can be obtained by the integral:

\[
\mathcal{h}(\mathbf{B}) = \int_{\mathcal{D}} \mathbf{A} \cdot \mathbf{B} \, d^3 r
\]

(1)

where \( \mathbf{A} \) is a vector potential of \( \mathbf{B} \). Since \( \mathbf{B} \) is the curl of the vector potential \( \mathbf{A} \) and defines the rotation of \( \mathbf{A} \) around a point, helicity indicates how much \( \mathbf{A} \) rotates around itself times its modulus. In an electromagnetic field in a vacuum, the electric field \( \mathbf{E} \) and magnetic field \( \mathbf{B} \) are divergenceless, and their vector potentials \( \mathbf{A} \) and \( \mathbf{C} \) are:

\[
\mathbf{E} = \nabla \times \mathbf{C} = -\frac{\partial \mathbf{A}}{\partial t}
\]

(2)

\[
\mathbf{B} = \nabla \times \mathbf{A} = -\frac{\partial \mathbf{C}}{\partial t}
\]

(3)

The helicity of the electromagnetic field is defined as:

\[
\mathcal{H} = \frac{1}{2} \int \mathbf{A} \times \mathbf{B} - \mathbf{C} \times \mathbf{E} \, d^3 r
\]

(4)

Here, the first and second terms are the magnetic field helicity \( \mathcal{H}_{\text{mag}} \) and the electric field helicity \( \mathcal{H}_{\text{elc}} \), respectively.[22]

\[
\mathcal{H}_{\text{dc}} = \frac{1}{2} \int (\mathbf{C} \cdot \mathbf{E}) \, d^3 r
\]

(5)

\[
\mathcal{H}_{\text{mag}} = \frac{1}{2} \int (\mathbf{A} \cdot \mathbf{B}) \, d^3 r
\]

(6)

\( \mathcal{H} \) is a conserved pseudoscalar, and it is gauge-invariant under the same conditions as \( \mathcal{H}_{\text{mag}} \) and \( \mathcal{H}_{\text{dc}} \).

From another aspect of particle physics, helicity is also defined as the projection of the angular momentum in the motion direction:

\[
\mathcal{H}' = \frac{\mathbf{p} \cdot \mathbf{J}}{\mathbf{p}}
\]

(7)

where \( \mathbf{p} \) is the momentum, and \( \mathbf{J} \) is the angular momentum, which is defined as \[^{22,23} \]

\[
\mathbf{J} = \epsilon_0 \int r \times (\mathbf{E} \times \mathbf{B}) \, d^3 r
\]

(8)

It is known that \( \mathbf{J} \) can be separated into two parts, that is, the orbital part \( \mathbf{L} \) and the spin part \( \mathbf{S} \).[24,25]

\[
\mathbf{L} = \epsilon_0 \oint \mathbf{r} \times (\mathbf{E} \times \mathbf{V}) \mathbf{A}_l
\]

(9)

\[
\mathbf{S} = \epsilon_0 \oint \mathbf{r} (\mathbf{E} \times \mathbf{A}_s)
\]

(10)

The spin angular momentum of chiral light fields is associated with circular polarization (CP), and the orbital angular momentum depends on the presence of helical phase fronts.[23]

In 1964, Lipkin introduced a conserved quantity of 00-Zilch for describing the helicity of light field:[26]

\[
\mathcal{Z}^{00} = \frac{1}{2} \int (\mathbf{E} \cdot \nabla \times \mathbf{E} + \mathbf{B} \cdot \nabla \times \mathbf{B}) \, d^3 r
\]

(11)

It is a higher-order extension of the optical helicity obtainable from the optical helicity by replacing the electric and magnetic fields with their curls.[23] However, Lipkin and others subsequently dismissed this quantity as having no physical significance until Tang et al. employed it to measure the chirality of light. This quantity is known as the optical chirality or the chirality density, that is, geometrically, the field lines wrap around a central axis with a component parallel to that axis.[1] Similar to the energy density and Poynting energy flow, the chirality density \( \mathcal{C} \) and the chirality flow \( \phi \) are expressed as:

\[
\mathcal{C} = \frac{\epsilon_0}{2} \mathbf{E} \cdot \nabla \times \mathbf{E} + \frac{1}{2 \mu_0} \mathbf{B} \cdot \nabla \times \mathbf{B} = -\frac{\epsilon_0 \omega}{2} \text{Im} (\mathbf{E}^* \cdot \mathbf{B})
\]

(12)

\[
\phi = \epsilon_0 \omega^2 \left[ (\mathbf{E} \times (\nabla \times \mathbf{B}) - \mathbf{B} \times (\nabla \times \mathbf{E}) \right]
\]

(13)

which also satisfy the continuity equation:[12,27]

\[
\frac{\partial \mathbf{C}}{\partial t} + \nabla \cdot \mathbf{E} = 0
\]

(14)
The interaction between chiral molecules and an electromagnetic field is described by the excitation rate. When a chiral molecule is subjected to a monochromatic electromagnetic field, it generates an electric dipole moment \( \mathbf{p} \) and a magnetic dipole moment \( \mathbf{m} \) defined as:

\[
\mathbf{p} = \hat{a} \mathbf{E} - i \hat{c} \mathbf{B}, \quad \mathbf{m} = \hat{c} \mathbf{B} + i \hat{a} \mathbf{E}
\]

where \( \hat{a} = a' + ia'' \) is the electric polarizability, \( \hat{c} = c' + ic'' \) is the magnetic susceptibility, and \( \hat{G} = G' + iG'' \) is the isotropic mixed electric-magnetic dipole polarizability. The excitation rate of the molecule by the field is:

\[
A^\pm = \langle \mathbf{E} \cdot \mathbf{p} + \mathbf{B} \cdot \mathbf{m} \rangle = \frac{\omega}{2} \left( a''|\mathbf{E}|^2 + c'|\mathbf{B}|^2 \right) \pm G'' \omega \text{Im} (\mathbf{E} \cdot \mathbf{B})
\]

where the brackets indicate an average over time. Since \( \chi \) is small for most molecules, the second term of Equation (16) can be ignored. By substituting \( \text{Im} (\mathbf{E} \cdot \mathbf{B}) = \mathbf{B} \cdot \mathbf{E} - \mathbf{E} \cdot \mathbf{B} \), the following is obtained:

\[
A^\pm = \frac{2}{\epsilon_0} \frac{\omega U_c a''}{2} \left( C G'' \right)
\]

where \( U_c = \frac{\omega}{4} |\mathbf{E}|^2 \) is the time-averaged electric energy density. The dissymmetry factor is calculated by \( g = 2(A^+ - A^-)/(A^+ + A^-) \); thus, \( g \) includes any pair of electromagnetic fields defined as:

\[
g = - \left( \frac{G''}{a''} \right) \left( \frac{2C}{\omega U_c} \right)
\]

Here, the first term in Equation (18) denotes the parameters of the chiral molecules, and the second term denotes those of the electromagnetic fields. The equation indicates that the chiral asymmetry of the excitation rate of the chiral molecules is proportional to the product of the chirality of the molecules and the electromagnetic fields. Large chiral asymmetry can be achieved by constructing a superchiral field with a large value of the second term in Equation (18). It is known that the dissymmetry factor excited by CPL is \( g_{CPL} = -4G''/ca'' \); thus, the increase in the dissymmetry factor by using a superchiral near-field compared to CPL can be evaluated by:

\[
g = \frac{g}{g_{CPL}} = \frac{cC}{2\omega U_c}
\]

3.1. Chiral Near-Fields in the Vicinity of 3D Chiral Structures

As the most intuitive chiral structure, helical structures have attracted much attention for generating chiral near-fields. When the rotational direction of a single helix matches the polarization of incident CPL, chiral near-field responses occur, as shown in Figure 6a. Meanwhile, the enhanced optical chirality of its enantiomers is symmetric.\[10\] For multiple helices, the chiral near-fields over an extended region are illuminated by the linearly polarized light, which is used to stimulate the resonance of the structure. The single-handed chiral near-fields in the helices are the results of the chiral eigenmodes of the structures, which could be used for discriminating enantiomers.\[11\] Besides helical nanostructures, 3D chiral oligomers composed of nanoparticles have drawn much interest due to the tunability and strong coupling effect between particles in the generation of superchiral fields.\[10,28,29\] In 2012, Schäferling et al. proposed a chiral oligomer composed of two twisted L-shaped layers.\[10\] Similar to the helix, strong chiral near-field responses occur with matching polarized light illumination. The strongest chirality enhancement is observed in the small gaps between two disks due to local electromagnetic field enhancement. In 2014, Ogier et al. fabricated chiral nanoparticle oligomers (Figure 1b) with facile hole-mask colloidal lithography to achieve strong chiral far-field and near-field responses.\[28\] Chiral hotspots with strong chiral near-fields (\( C_{CPL} \approx 200 – 300 \)) occur in the gap between nanoparticles, corresponding to the “hot spots” of electromagnetic fields. The average optical chirality surrounding the oligomers is \( \pm 5 \) at the plasmonic resonance peak. This method may have potential for the detection and analysis of a small number of molecules.

3.2. Chiral Near-Fields in the Vicinity of 2D Chiral Structures

However, the preparation of 3D chiral structures is relatively complex. It has been demonstrated that planar chiral structures could be used to generate chiral near-fields.\[9,10,30-33\] The gamma- dion structure, a classic planar chiral structure, has been widely studied theoretically and experimentally.\[9,10,32\] In 2012, Schäferling et al. demonstrated that the regions of the highest enhancement of the chiral near-fields and electric fields in the gamma- dion structure are not identical, indicating that strong electric fields are not sufficient to produce superchiral fields.\[10\] Further, they demonstrated that the superchiral field of the magmomd structure is insensitive to the polarization state of incident CPL and the enhanced optical chirality is discontinuous in space and quite small. To overcome these disadvantages, a two-armed nanospi- ral was proposed to improve the chiral near-field properties (Figure 1c).\[10\] As expected, the chiral near-fields of the nanospi- rals have smoother distributions and are different for two incident polarizations; two opposite chiral near-fields occur on either side of the structure. This approach could have potential for enantiomer sensing and optical rotation measurements. In 2017, Kang et al. achieved the precise control of chiral near-fields using “superchiral light,” including chiral switching and selective enhancement.\[30\] In addition, the superchiral fields are more de-localized, which improves the potential for nanostructure-based enantiomeric sensing. Furthermore, simpler structures such as staggered nanorod dimer\[9\] and nanoslices\[31\] arrays were theore-
Figure 1. Chiral near-fields generated by 3D and 2D chiral nanostructures. a) Optical chirality enhancement of helixes excited by matching handed circularly polarized light (CPL). Reproduced under the terms of the Creative Commons Attribution 3.0 License.[10] Copyright 2012, The American Physical Society. b) Schematic of chiral nanoparticle oligomers excited by CPL. Reproduced with permission.[28] Copyright 2014, The American Chemical Society. c) Optical chirality enhancement of two-armed nanospirals under left-handed CPL (LCP) (left) and right-handed CPL (RCP) illuminations (right). Reproduced under the terms of the Creative Commons Attribution 3.0 License.[10] Copyright 2012, The American Physical Society. d) Schematic of chiral near-fields generated with a nanorod (left top), a nanoslit (middle top), and nanoslit dimers (left middle), and the chiral near-field distributions of the nanoslit dimer (right). Reproduced with permission.[31] Copyright 2012, The American Chemical Society.

3.3. Chiral Near-Fields in the Vicinity of Achiral Structures with Linear Polarization

Besides chiral nanostructures that are used to generate chiral near-fields, local chiral fields have also been observed near symmetric nanostructures under certain conditions.[12,13,34,35] This response is called “extrinsic chirality.” In 2012, the Schäferling group first theoretically demonstrated the existence of “extrinsic chirality” near-fields that could be generated with achiral nanostructures under linearly polarized light.[12,34] As shown in Figure 2a, chiral near-fields appear near a nanoantenna under normal incidence and are caused by the interference between incident and scattered fields.[12] The authors explained the phenomenon using a simple dipole model. Similarly, the Davis group also independently obtained chiral near-field enhancements using an achiral structure composed of three nanorods in 2013.[34] Uniform and strong superchiral fields are crucial in many applications. Based on this, Tian et al. designed a simple Au dimer structure and excited it with linearly polarized light in the diagonal direction of the structure,[13] as depicted in Figure 2b. Simulation results showed that one-handed chiral near-fields and electric fields were significantly enhanced in the gap of dimers, and the handedness of the chiral near-fields could be inverted by changing the incident polarization to the other diagonal direction of the dimer. The volume-averaged chiral field enhancement in the whole gaps was 30-fold, exhibiting the potential for chiral sensing and Raman optical activity (ROA) measurements of small quantities of chiral molecules. The authors explained the mechanisms using an analytical dipole model. On this basis, Hu et al. enhanced the local chiral field by adding an Au film under the block dimer.[36] Further, they used a block heterodimer with...
different sizes on the Au film and achieved one-handed chiral near-fields with opposite chirality in different regions.\(^{[37]}\)

### 3.4. Hidden Chirality

Different from “extrinsic chirality” with off-resonance, “hidden chirality” was obtained in achiral nanosystems with symmetrical stimulations\(^{[38–40]}\) resulting from the interference between the two plasmonic eigenmodes excited by CPL. In 2018, Zu et al. acquired an image and controlled the chiral electromagnetic modes of the V-shaped nanostructure with CP-resolved cathodoluminescence (CL) microscopy (Figure 2c).\(^{[38]}\) The chiral electromagnetic modes and chiral radiative local density of nanostructures’ states were observed in the CL image. The chirality of the symmetric nanostructure has an extreme chiral distribution (\(\approx 99\%\)) at the arm ends, which can be used to detect polarized CL emission from the hotspot and determine the radiative local density of states. Subsequently, Horrer et al. achieved symmetric local optical chirality with a symmetric trimer consisting of three gold nanodisks\(^{[39]}\) under LCP and RCP, as shown in Figure 2d. They experimentally demonstrated that the optical chirality could be imprinted onto the photosensitive polymer, which could be used in polarization-sensitive photochemistry.

### 4. Chiral Near-Fields Generated by Photonic Enhancement

As discussed above, various plasmonic nanostructures composed of metallic elements have been designed to obtain strong chiral near-fields due to highly enhanced local electromagnetic fields. Equation (12) indicates the importance of the magnetic field in generating chiral near-fields. However, the magnetic modes of metal structures are either too weak or separate from the electric modes, resulting in challenges in achieving strong and one-handed chiral near-fields. Some negative-index metamaterials and all-dielectric nanostructures have been utilized to improve the design and obtain chiral near-fields.

Since negative-index materials can generate strong local magnetic fields, Yoo et al. proposed an achiral negative-index metamaterial composed of stacked double-fishnet layers (Figure 3a)\(^{[41]}\) to obtain highly enhanced chiral near-fields.\(^{[41]}\) Large-volume and one-handed chiral near-fields were generated in the entire volume inside each cavity, which is desirable for chiral molecule sensing. Further, it was demonstrated that the volume-averaged optical chirality could be further increased by changing the number of stacked layers. The results offer a new approach for enhancing chiral near-fields with metamaterials.
In addition to negative-index materials, all-dielectric metamaterials have recently attracted much attention due to their low heat generation, low intrinsic loss, high-quality factors, and high magnetic resonance. Based on these properties, numerous studies were conducted on these dielectric materials to obtain unique electromagnetic properties. Ho et al. demonstrated that magnetic multipolar resonances of sub-micron silicon spheres were excited by CPL. The maximum optical chirality and Kuhn’s dissymmetry factor were observed near the magnetic resonances. Solomon et al. theoretically investigated how to design large area, uniform sign chiral near-fields with high index dielectric metasurfaces. They used metasurfaces composed of silicon disks and obtained a 138-fold enhancement of optical chirality and a 30-fold enhancement of volume-averaged optical chirality near the disks. By tuning the aspect ratio of the disks, the maximum one-handed local chiral field was achieved when the electric and magnetic modes had the same frequency (Figure 3b) due to a spatial overlap of the electric and magnetic fields.

Mohammadi et al. considered similar design rules and obtained highly enhanced chiral near-fields with dielectric nanoparticles with the synergistic combination of electric and magnetic resonances. A dielectric dimer composed of two silicon spheres was used (Figure 3c), and parallel, spatially overlapping, and ideal phase retardation (π/2) of the electric and magnetic fields were generated in the hot spots. Holey silicon disk resonators were proposed based on the K erker effect to simplify the structure. Uniform chiral near-fields were achieved by the periodic resonators. Furthermore, Du et al. used a single hollow silicon disk (Figure 3d) and systematically analyzed the relationship between the dipolar interference and chiral near-fields using the multipole decomposition method. The results showed that the maximum enhancement of the averaged optical chirality is related to the magnetic dipole and anapole resonances. Meanwhile, the optical chirality enhancement could be tuned by size scaling. This research provided a potential method for chirality detection with all-dielectric metamaterials.

5. Ultra-Limit Detection with Chiral Near-Fields

Due to their excellent photoelectric properties, chiral near-fields have potential applications in molecular detection, recognition, separation, and sensing. Since the chiral response of natural chiral molecules is very weak, the detection sample is limited to the microgram level. To detect the chiral molecules at the picogram level, superchiral fields generated by plasmonic metamaterials have been introduced. Hendry et al. first experimentally demonstrated that superchiral fields could be generated in planar gammadion-shaped nanostructures, as shown in Figure 4a. When chiral analytes were adsorbed to the chiral nanostructures, wavelength shifts were observed as chiral responses of the nanostructures; they depend on the handedness of the chiral molecules (Figure 4a-i). The authors utilized the dissymmetry factor (g = nR − nL / nR + nL, and nR and nL are the effective refractive indices of the chiral molecules for the right-/left-handed nanostructures, respectively) to measure the strength of interaction between the chiral molecules and the nanostructures. It was 106 times greater than that of the same molecules in solution with...
The authors further verified that the strong chiral response was attributed to an increase in the local fields and the superchiral fields caused by the coupling between the different branches of the nanostructures, as shown in Figure 4a-ii.

From another perspective, Tang et al. proposed enantioselective excitation of the chiral molecules with superchiral light, produced by the superposition of CPL with opposite chirality and slightly different amplitudes propagating in opposite directions. When the chiral fluorescent compounds were excited by the superchirallight in the experiment, the dissymmetry factors were 11 times greater than those obtained by CPL, and their signs were the opposite for the enantiomers. They pointed out that the enhancement of the dissymmetry factors resulted from the suppression of electric energy density at the nodes. The enantioselective excitation could be carried out for any small chiral molecule.

Ultra-limit detection of high-order structures using conventional chiroptical spectroscopic method is challenging because their signal is less sensitive to change than that of lower-order structures. However, the superchiral evanescent near-fields of the chiral nanostructures are sensitive to higher-order structures. Based on this, Tullius et al. demonstrated that the higher-order structures of biomolecules could be detected by new label-free biophysical measurements using the superchiral evanescent near-fields of “shuriken” nanostructures (Figure 4b).

It is well known that chiral plasmonic nanostructures could produce large CD signals, which are stronger than those of chiral molecules, preventing the sensitive detection of the molecular signals. To solve this problem, Garcia-Guirado et al. developed a plasmonic sensor consisting of racemic plasmonic arrays of 2D chiral structures, which showed strong chiral near-field responses but no far-field CD responses (Figure 4c). However, the CD signal appears when in contact with a chiral molecular layer. This sensor was used to distinguish the handedness of phenylalanine molecules. Similar racemic nanograting was also used for detecting chiral molecules.

As discussed above, chiral near-fields have a wide range of applications in detection, sensing, and related fields. Therefore, it is important to obtain accurate measurements of chiral near-fields around nanostructures. Meinzer et al. experimentally analyzed a chiral near-field using luminescence enhancement of achiral dye molecules around chiral nanostructures. As shown in Figure 4d, the dissymmetry of the luminescence enhancement is correlated with the optical chirality. This method can accurately detect a chiral near-field around the nanostructure, even with a very weak CD response.

Moreover, strong and uniform chiral near-fields are required to detect a small quantity of molecules or a single molecule. Nanostructures producing chiral near-fields should have simple configurations to be fabricated conveniently and economically. In addition, it is preferable to use linearly polarized light rather than CPL, which requires a broadband circular polarizer. Theoretical research has been conducted to meet these requirements. In practice, molecules are typically randomly distributed around a nanostructure or in a specific area. In 2015, Finazzi et al. pointed out that the volume-averaged optical chirality around small plas-
monic nanoparticles could not be larger than that in a plane wave due to the quasistatic nature. The volume-averaged optical chirality may be stronger near larger nanostructures if the signs of the local optical chirality do not change. It was found that one-handed chiral near-fields could be obtained by adding a layer to cover the two opposite corners of squares to prevent cancellation between the chiral near-fields with different handedness in a nanoantenna application. Thus, the molecules could be placed in the groove and interact with the one-handed chiral near-fields. As mentioned in Section 3, strong uniform chiral near-fields could be produced by a helical spring structure. Based on this, Schäferling et al. designed a simplified nanostructure composed of a diagonal slit array in a metallic film on a mirror to minimize fabrication difficulty. The properties of the chiral near-fields could be precisely controlled by the distance between the two layers, and one-handed chiral fields could be obtained if the distance was sufficiently small. In experiments, chiral molecules could be placed in the slits and analyzed using chiroptical spectroscopy. Meanwhile, the detection of ROA of a single molecule or a few molecules has attracted significant attention. This process is difficult due to the small cross section. Hu et al. proposed a theoretical equation to calculate the ROA enhancement factor similar to that of surface-enhanced Raman scattering (SERS). They discussed the ultimate limit of SE-ROA. With the structure consisting of Au block dimer on an Au film, SERS and ROA were highly enhanced in the gap, providing a theoretical basis for the ROA detection of a single molecule in experiments.

However, SE-ROA has several drawbacks, such as high photothermal heat and an inability to transfer and enhance the optical chirality from the far field to the near-field. Therefore, Xiao et al. theoretically and experimentally demonstrated that the chiral-field enhanced ROA could be significantly increased by tuning the chiral near-field with a silicon nanodisk array in the dark mode. The average enhancement factor was $\approx 10^2$ in the near-field region.

6. Spin-Orbit Interaction in Chiral Fields

As discussed in Section 2, photons have angular momentum, that is, spin angular momentum and orbital angular momentum. Opposite spin angular momenta correspond to...
RCP and LCP, whereas the orbital angular momentum is associated with the direction of the wave vector and the phase fronts. The corresponding canonical momentum densities of the field are $P_o = \frac{1}{2} \text{Im} [\varepsilon \mathbf{E} \cdot (\nabla) \mathbf{E} + \mu \mathbf{H} \cdot (\nabla) \mathbf{H}]$ and $S_o = \frac{1}{2} \text{Im} [\varepsilon \mathbf{E} \times \mathbf{E} + \mu \mathbf{H} \times \mathbf{H}]$ where $g = \frac{(8\pi\omega)^{-1}}{\eta}$. When light interacts with materials with an inhomogeneous refraction index, the conservation of the optical angular momentum leads to a momentum transfer between the spin (polarization) and orbital (propagation and phase) momenta. As a result, a change in the light’s trajectory accompanying a transverse spin-split of the beam occurs. This phenomenon is the photonic spin Hall effect (PSHE), a counterpart of the well-known electronic SHE, where the electrons spin-dependently accumulate at opposite sides of an electron flow due to the spin-orbit interaction.

In conventional cases, the photonic spin-orbit interaction (PSOI) is typically very weak for a small interaction volume. Special techniques are required to observe this phenomenon, such as weak quantum measurements or multiple total internal reflections, limiting its application. Metal nanostructures provide a reliable approach to achieving strong light–matter interaction due to their plasmonic effects. Moreover, a large gradient of permittivity exists on the metal-dielectric interface, resulting in strong PSOI. Yin et al. utilized a metasurface with V-shaped gold antennas to obtain strong PSOI. They directly observed a strong PSHE with a large opposite transverse motion of RCP and LCP represented by the red and blue areas (the color represents the chiral field C) in Figure 6a. Subsequently, researchers developed different types of novel spin Hall devices, such as a spin-dependent splitter, Stokes parameter detector, and the Pancharatnam–Berry phase lens, by manipulating the PSOI with subtly designed metasurfaces.

Light with both spin and orbital angular momentum can be tailored by the PSOI from the incident light with only spin angular momentum. The interaction between this type of light (Laguerre–Gaussian (LG) beam) and plasmonic nanostructures has been investigated in recent years. Orbital angular momentu-
7. Chiral Field for Optical Trapping

Light has linear momentum, which can be transferred to particles via light-matter interaction, pushing the particle forward in the direction of the wave vector. This phenomenon is known as the radiation force, which can be used to manipulate particles.\(^{[58]}\) When a particle is located in the narrow beam of a laser, the gradient force occurs due to spatial inhomogeneity of intensity. This force has been widely used to power optical tweezers,\(^{[59]}\) and is seen as a promising method to interact with chiral molecules due to the effective and noninvasive interaction between chiral light and chiral objects.\(^{[60]}\) It was discovered in the last decade that the lateral force separates chiral objects. It moves objects with different chiralities in opposite directions. The lateral force arises from the transverse spin angular momentum of the field and can be generated by placing an object above a slab, in an evanescent wave, or in the interference light field.\(^{[58,60,61]}\)

Recently, Li et al. demonstrated the sorting of chiral particles by a tightly focused beam.\(^{[62]}\) If a dipolar particle is much smaller than the wavelength, the optical force within the dipole approximation can be analytically expressed as:

\[
\langle F \rangle = \nabla \langle U \rangle + \frac{\sigma_m}{c} \langle S \rangle - i \text{Im} \left[ a_m \right] \nabla \times \langle S \rangle - \frac{c_0}{n_1} \nabla \times \langle L_m \rangle - \omega r_m \langle L_m \rangle + \text{Im} \left[ a_m \right] \nabla \times \langle L_m \rangle
\]

\[
+ \frac{ck^2}{12c n_1} \text{Im} \left[ a_m a_m^* \right] \text{Im} \left[ E \times H \right]
\]

with the optical potential\(^{[62]}\)

\[
\langle U \rangle = \frac{1}{4} \text{Re} \left[ a_m \right] |E|^2 + \frac{1}{4} \text{Re} \left[ a_m \right] |H|^2 + \frac{1}{2} \text{Re} \left[ a_m \right] \text{Im} \left[ E \cdot H \right]
\]

where \(E\) and \(H\) are the electric and magnetic fields acting on the particle, \(\langle S \rangle\) is the time-averaged Poynting vector, \(\langle L_m \rangle\) and \(\langle L_m \rangle\) are respectively the electric and magnetic parts of the time-averaged spin angular momentum density, \(\text{Im} \left[ E \cdot H \right]\) is the chiral near-field, and \(n_1\) is the refractive index of the medium with relative permittivity \(\varepsilon_1\) and permeability in a vacuum; \(\sigma_m, a_m, a_m^*\) are respectively the light speed, permittivity, and permeability in a vacuum; \(\sigma_m, a_m, a_m^*\) are respectively the electric, magnetic, and chiral polarizabilities of the chiral particle, respectively.

The PSOI induces a transformation of the spin and orbital angular momenta of the light transfer to the electron resonance of LSPs with the requisite of quadrupole or higher multipolarity excitation.\(^{[57]}\) The PSOI allows a transformation of the spin and orbital angular momenta. Due to the spatial distribution of the phase front, the isophase surface of the LG beam (with nonzero orbital angular momentum) will sweep over the nanostructure over time. With the involvement of both spin and orbital angular momenta by PSOI, the partial spatial excitation of the plasmonic nanostructure generates strong dichroism that is two orders of magnitude higher than that excited by light with only spin angular momentum (CPL). The plasmon modes excited by light with orbital angular momentum also have significantly different charge distributions and superchiral near-fields (Figure 6b). When the incident light has spin and orbital angular momenta with the same sign, the superchiral near-fields of the excited plasmon modes are more enhanced than those excited by light without orbital angular momentum (considering the very low intensity of the central area of the LG beam). Meanwhile, when the spin and orbital angular momenta have opposite signs, they cancel the superchiral near-field.\(^{[56]}\) The PSOI can result in a significant enhancement of the superchiral near-field under the right conditions of the spin and orbital momenta.

8. Discussion and Outlook

Plasmon-enhanced chiral near-fields can match chiral molecules and light wavelengths with large size differences, significantly enhancing the CD of the chiral molecules. Plasmonic nanostructures have dimensions of tens to hundreds of nanometers, which is much larger than that of chiral molecules. Thus, it is necessary to develop nanostructures with higher chirality density to achieve higher CD enhancement of the chiral molecules. The fabrication of plasmonic chiral structures with a size of several nanometers or smaller is challenging using current nanofabrication techniques. The plasmonic nanogap enables the generation of a highly distorted chiral near-field. However, the small gap size limits the application of this method. The design and fabrication of plasmonic nanostructures to create superchiral near-fields with high chirality density are important research topics.

A crucial aspect of research on the plasmon-enhanced chiral near-field is its distribution. However, the detection of the near-field distribution is challenging due to a lack of convenient and...
Figure 7. a) Transverse optical force distributions of particles with different chirality parameters. b) The radial (left) and azimuthal (right) optical forces as functions of the particle’s radial displacement and chirality parameter. The inset is a line scan of the radial optical force with $\chi = -0.38$ in the radial direction. c) The contributions of the decomposed radial (left) and azimuthal (right) optical forces with $\chi = 0.5 + 0.5i$. (a–c) Reproduced with permission. Copyright 2019, The American Physical Society. d) The contributions of the decomposed gradient forces with $\rho = 0.1 \mu m$.

widely used far-field methods. Luminescence enhancement with dye molecules or quantum dots and leakage radiation microscopy are typically used to image the near-field distribution of plasmon waves.\cite{52,63} However, these methods are not applicable to the detection of plasmon-enhanced chiral near-fields due to the low resolution. At present, the most effective method to achieve this goal is scanning near-field optical microscopy.\cite{52} It precisely images the amplitude distribution of the chiral near-field and provides phase information. Nevertheless, the setup is quite complex and difficult to use, making the detection of the plasmon-enhanced chiral near-field challenging.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

chiral nanostructures, circular dichroism, near-field, superchiral fields, surface plasmons

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