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Enhancing circular dichroism by super chiral hot spots from a chiral metasurface with apexes

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Manipulating light spin (or circular polarization) is an important research field and may find broad applications from sensors, display technology, to quantum computing and communication. To this end, planar metasurfaces with larger circular dichroism are strongly demanded. However, current planar chiral metasurface structures suffer from either fabrication challenge, especially from near-infrared to visible spectrum, or insufficient circular dichroism. Here, we report a chiral metasurface composed of achiral nanoholes which allow for precisely creating apexes in the designed structure. Our investigation indicates that the apexes act as super chiral hot spots and enable the highly concentrated near-field optical chirality leading to a remarkable enhancement of circular dichroism in the far-field. A 4-fold enhancement of the circular dichroism and a strong optical activity of ~ 15 degrees have been experimentally achieved. Besides the enhanced chirality, our design genuinely overcomes the nanofabrication challenge faced in existing planar chiral metasurfaces.

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Chirality is a ubiquitous property in nature, from galaxies in universe to amino acids in life science, and have been attracting intense studies in recent decades because of its extreme importance in the field of modern photonics. Structural chiral materials can usually appear in two forms called enantiomers, which are geometrical mirror images of each other and are often associated with very different biological functions. In the realm of optics, chiral media shows different optical responses, when excited by circular polarized light (CPL), with different spin states, and give rise to significant optical effects, such as optical activity (OA) and circular dichroism (CD). However, for naturally occurring chiral materials, the chiral responses are incredibly weak due to the mismatching between the much smaller molecular chiral structures and the “twist” of the CPL, which consequently causes a weak chiral matter-light interaction.¹

With the advent of metamaterials in the recent years, solutions have been sought through the design of artificial chiral building blocks:^{2–9} three dimensional (3D) chiral plasmonic metamaterials^{6,10–16} have not only exhibited tremendous enhanced chiral responses but also shown the ability of manipulating the light spin modes, which contributes to substantial benefits for various applications, such as ultrasensitive bio-detection,¹⁷ miniature circular polarizers,¹⁸ and quantum communications.¹⁹ However, despite significant innovation in nano-fabrication techniques,^{15,20} their realizations remain both time-consuming and expensive due to their structural complexity, especially for those working in the visible range.

With a view to tackle the above-mentioned problems, two dimensional (2D) chiral metamaterials, also known as

chiral metasurfaces,^{2,3,7,21–26} have been proposed to show remarkable chiral responses. Theoretical calculations^{24,25} and experiments²⁶ have proved that this surprising effect relies on circular polarization dependent finite non-radiative (Ohmic) losses of the metasurfaces. This means that the planar chiral structures can break the in-plane mirror-symmetry and thus can generate the asymmetric distributions of localized or propagating plasmon modes^{22,23,27} corresponding to different spin states, which results in CD. Notwithstanding the many examples of 2D chiral metasurfaces designs, such as gammadion shape^{3,28} and L-shape antennas,⁷ there are still very limited studies on the design methodology of such metasurfaces to obtain significant enhancement of the chiral response. Especially, most of the 2D chiral metasurfaces are composed of bars or slits containing sharp corners, which usually become obtuse during fabrication and thus compromise the actual performance.

Here, we propose a chiral metasurface, composed of an array of achiral nanoholes milled by an advanced focused ion beam (FIB) technique in an ultra-thin gold film. We demonstrate a straightforward design strategy that apexes can be readily formed in a metasurface by a slight overlap of the nanoholes. The optical chirality in the near-field is highly concentrated around the apexes due to the chiral-like interference between the achiral nanoholes so as to generate the super chiral hot spots. Experimentally, the designed structure well coincides with the FIB milling technique, which greatly mediates the fabrication difficulty usually exist in other approaches. As a result, a remarkable ~ 4 -fold enhancement of the circular dichroism has been achieved with the apexes added. A huge optical activity (OA) signal of $\sim 15^\circ$ is observed experimentally. Our results highlight the impact of

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FIG. 1. Schematic illustration of the investigated chiral metasurface (a) without (b) and with overlapping nanoholes (c) and (d) corresponding SEM images.

apexes on enlarging the chiral response of metasurfaces by generating the super chiral hot spots, potentially shedding a light on the design of next generation chiral metamaterials.

Sharp plasmonic structures,²⁹ such as the plasmonic bowtie antenna,^{30,31} have been widely studied and utilized to confine the optical fields and to enhance the near-field intensity. The choice of nanoholes as basic units of our metasurface allows us to easily either create or remove apexes by controlling the distance between the nanoholes without altering the rotatory feature of the chiral pattern. On the other hand, with the rich modes of achiral nanoholes, it is also possible for the tailoring of chiral responses.

As shown in Fig. 1, the chiral metamolecule is formed by 7 achiral nanoholes of identical diameter $a = 200$ nm, separated by a gap g that can span from positive [Fig. 1(a)] to negative [Fig. 1(b)] values: as a whole, this pattern resembles a rotated letter “Z” with an angle of 60° between the adjacent arms and possesses intrinsic chirality due to the breaking of in-plane mirror symmetry. The chiral cells are arranged in a square lattice array with a period p . Figures 1(c) and 1(d)

show scanning electron microscopy (SEM) images of the chiral metasurface, for values of the gap g of $g \sim 20$ nm (no overlap) and $g \sim -20$ nm (overlap), respectively, milled in a ~ 100 nm thin gold film on a glass substrate.

In order to investigate the resonant modes of chiral responses, we perform full-wave numerical simulations for three chiral patterns of varying gap size between the nanoholes: $g = 20$ nm (non-overlap nanoholes), $g = 0$ nm (touching nanoholes), and $g = -20$ nm (overlapping nanoholes). In the simulations, the chiral metasurfaces are excited at normal incidence, and the spectra of the circular dichroism (CD) are calculated by the differential transmittance between the left and right circular polarized light [Fig. 1(a)]: a huge chiral optical response is expected at the peak of the black curve, corresponding to the $g = -20$ nm, where the curve exceeds 10 percent. More importantly, from the three curves, it appears a trend of increasing chiral optical responses with the appearance of the apexes when the gaps start vanishing (red to green to black). The slight red shift of the peak with decreasing gap size can be understood by the fact that with smaller gaps, the mutual distance between adjacent metamolecules increases while the period remains constant so that the collective response of the metasurface, governed by the interference among metamolecules, is red-shifted. A clearer understanding of the effect can be drawn by plotting the electric field distribution maps of the three structures, at resonance [838 nm for Figs. 2(b) and 2(c); 850 nm for Figs. 2(d) and 2(e); 860 nm for Figs. 2(f) and 2(g)], for both left (LCP) and right (RCP) circular polarized light excitations, as shown in Figs. 2(b)–2(g). A comparison of the left [Figs. 2(b), 2(d), and 2(f)] and the right [Figs. 2(c), 2(e), and 2(g)] panels shows that the distributions of the field enhancement hot spots have a clear dependence on the circular polarizations of the incident light, accounting for the dramatic asymmetric transmittance of RCP and LCP of Fig. 2(a). Moreover, it can be observed that, once the apexes are generated, the intensity of the electric field hot spots located at the apexes’ area is highly enhanced (nearly one order of magnitude), which expressively links the electric field enhancement to the chiral response, which is not always

FIG. 2. (a) Simulated differential transmittance for the three samples, corresponding to the gap distances of 20 nm (red curve), 0 (green curve), and -20 nm (black curve): the peak value increases with the generation of the apexes. Simulated electric field distribution maps excited by right and left CPL at resonant wavelengths (838 nm for $g = 20$ nm; 850 nm for $g = 0$; and 860 nm for $g = -20$ nm) for the chiral patterns with the identical lattice period 800 nm and various gap distances of (b) and (c) $g = 20$ nm, (d) and (e) $g = 0$, (f) and (g) $g = -20$ nm, respectively.

a straightforward connection, as indicated in previous studies.³² In order to explicitly understand the physical mechanism behind, we can simply refer to the formula which describes the non-radiative (Ohmic) losses: $P_{abs} = \frac{1}{2} \omega \varepsilon'' |E|^2$, where the angular frequency ω and the imaginary part of the permittivity ε'' are constant. Obviously, when the electric field intensity $|E|$ dramatically increases with the apex generated, as shown in Figs. 2(b)–2(g), the asymmetric non-radiative (Ohmic) losses will become much larger because of the circular polarization dependent electric distribution; as a result, the metasurfaces with apexes show enhanced chiral responses.

To further elaborate this relationship, we quantitatively introduce the optical chirality C , a time-even pseudoscalar which is used to describe the spiral level of electromagnetic field:^{1,33,34} $C = -\frac{\varepsilon_0 \omega}{2} \text{Im}[\mathbf{E}^* \cdot \mathbf{B}]$, where the $\mathbf{E}$ and $\mathbf{B}$ denote the complex amplitudes of electric and magnetic fields, respectively. In contrast to the optical chirality of circular polarized light, chiral metamaterials enable several orders of magnitude higher intensity in the near field,^{31,35,36} which in the other hand contributes to the far field effects of tremendous circular dichroism or optical activity. Based on the results reported in the Fig. 2, we calculate the near-field optical chirality for our chiral metasurfaces with and without apexes and compare them in Fig. 3. For a clear demonstration, we normalized the optical chirality maps in the same scaling. Apparently, the optical chiral fields shown in Figs. 3(b) and 3(c) are highly enhanced and concentrated around the apexes, compared to the case of non-overlapping holes in Fig. 3(a). The role of asymmetric interference (constructive or destructive) among the unit cells should also be understood, because, besides the localized modes, propagating modes play a crucial role^{22,23} especially for the chiral metasurfaces milled on plasmonic metals, thus contributing to the collective chiral responses. It is understandable that chiral geometries with apexes more likely become super chiral hot spots when the condition of interference is satisfied, if compared with geometries presenting only smooth boundaries. We can therefore conclude that the presence of apexes in our

chiral design does not only produce an enhancement of the electric near-field but also concurrently increases the intensity of optical chirality C thus reinforcing the optical chiral response of the metasurface [supplementary material](#).

We verified our proposed design strategy by experimental measurements on two $50 \mu\text{m} \times 50 \mu\text{m}$ array samples fabricated by focused ion beam milling on a 100 nm thin gold sample deposited over a glass substrate. We selectively fabricated the patterns with gaps of 20 nm and -20 nm to illustrate the impact of the apexes on the chiral responses, while the touching holes with zero gap is left out, due to the extreme degree of difficulty in fabrication. We determined the CD spectra by measuring normal incidence transmittance of both left and right circular polarized light using a commercial microspectrophotometer (Jasco MSV-5200) coupled with broadband quarter waveplate: the CD signal was deduced in terms of the ellipticity θ by the expression: $\tan \theta = \frac{\sqrt{T_{RCP}} - \sqrt{T_{LCP}}}{\sqrt{T_{RCP}} + \sqrt{T_{LCP}}}$. The numerical and experimental results displayed in Fig. 4 show remarkable agreement; within the fabrication accuracy, they both display a 4-fold enhancement. Furthermore, the black curve in Fig. 4(b), reaches an outstanding CD peak value of 2.1° that becomes comparable to the chiral responses of three dimensional chiral metamaterials.^{10–16} The blue shift of the experimental results corresponding to the simulation results could be explained by the fabrication inaccuracy. It is understandable that the actual diameter of nanoholes because of the ion etching effect will be slightly larger than that in simulations, which is equivalent to decrease the distances between the chiral metamolecules, thus resulting in the blue shift of the collective chiral responses in experiments.

Finally, we investigate the role of asymmetric interference generated by the above-mentioned propagating mode, which allows an easy spectral tuning of the chiral hot spots resonance wavelength by altering the lattice period: this resembles a cavity modifying the interference between the chiral metamolecules. Experimentally, we fabricated chiral metasurfaces with overlapping nanoholes of various periods ($p = 770, 800$, and 830 nm) but identical unit cell and

FIG. 3. Simulated field distribution maps of optical chirality excited by right and left CPL at the resonant wavelengths (838 nm for $g = 20$ nm; 850 nm for $g = 0$; 860 nm for $g = -20$ nm) for the chiral patterns with identical lattice period 800 nm and various gap distances of (a) $g = 20$ nm, (b) $g = 0$, (c) $g = -20$ nm, respectively.

FIG. 4. (a) Simulated and (b) experimental circular dichroism spectra corresponding to the chiral metasurfaces with (black curve) or without (red curve) apices.

FIG. 5. Experimental optical activity spectra (a) and circular dichroism spectra (b) corresponding to the chiral metasurfaces with the identical unit cell but various periods.

measured both their optical activity and circular dichroism spectra, as shown in Fig. 5. The peaks of both OA and CD spectra are significantly red-shifted proportionally to the increment of the period, which visibly confirms the modulation of the propagating mode on the super chiral hot spots. Moreover, for a period of 830 nm, the OA spectrum reaches a peak value of 15 degrees which makes our proposed chiral metasurfaces suitable for applications in ultra-compact optical rotators.

In conclusion, we have proposed a chiral metasurface composed of achiral nanoholes which allow to create apices in the nanostructured design and have investigated the impact of the apices on the metamaterial chiral response. We found that the chiral metasurface with apices give a greatly enhanced optical chiral responses. We demonstrated that the presence of the apices highly enhances and concentrates the intensity of both electric field and optical chirality in the near field and reported a 15 degree optical activity and large circular dichroism in planar metasurfaces. These super

chiral hot spots allow more efficient chiral-matter interactions which could potentially benefit in various applications. We suggest that the inclusion of apices in metamaterial designs could prove as a practical strategy to enhance their chiral optical responses and should be investigated further.

See [supplementary material](#) for detail information of fabrication, experimental measurements, and numerical simulation.

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