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A Novel Chiral Metasurface With Controllable Circular Dichroism Induced by Coupling Localized and Propagating Modes

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Abstract: The enhanced chiral response exhibited by chiral metamaterials and metasurfaces is generally attributed to the effect of localized surface plasmon resonances which are mostly determined by the constituent materials and structure geometry. How the presence of propagating surface plasmon modes influences chirality is still short of investigation. Herein, we introduce a novel chiral metasurface composed of a chiral arrangement of nanoslits carved in a continuous metal film, in which propagating surface plasmon modes are proved to be powerful tools to control the metasurface's chiral response. Numerical simulations help us devise a straightforward approach to the metasurface's design and through which, by modifying the coupling between localized and propagating modes, we experimentally achieved controllable and remarkably strong chiral responses. Our finding provides an extra degree of freedom in manipulating chiral responses, which may aid in the further design of chiral plasmonic structures.

Chirality is a key molecular structural concept and a ubiquitous phenomenon in nature which has become an increasingly significant research avenue since it was introduced in the context of metamaterials^[1, 2]. Compared with the weak chiral optical response of the natural chiral media, carefully engineered chiral metamaterials structures exhibit orders of magnitude larger chiral effects, thus making chiral metamaterials highly promising candidates for a wide range of applications^[3], including ultrasensitive chiral biomolecule sensing^[4-7], ultrathin broadband circular polarizer devices^[8-12] and negative refractive index materials^[13-16]. In recent years, planar metamaterials and metasurfaces^[17-20] have garnered intense attentions thanks to both their unique ability of manipulating light and the lower complexity required in fabrication because, despite the incredible recent progresses of nanofabrication technologies, three-dimensional chiral metamaterials^[21-27] while showing outstanding performances, remain both time-consuming and expensive to manufacture. Therefore, with a view to achieving practical photonic devices, planar metamaterials and metasurfaces are deemed a more promising platform because of fabrication simplicity and compactness for on-chip integration.

The research on chiral metasurfaces has covered an astoundingly large portfolio of designs, ranging from the initial famous gammadion shape meta-atom^[16, 28] to split-rings^[29-31], fish-shapes^[32], L-shape^[33], and achiral oriented structures^[34-37], and spanned the frequencies from visible to gigahertz(GHz)^[38]. For the purpose of chiral molecule sensing^[39, 40], it is essential to achieve a pronounced chiral optical response in the UV to near IR spectral range, where the resonant responses of most chiral molecules lie. However, the existing planar structures either exhibit a weak chiral response or require illuminations at quite shallow angles of incidence. More importantly, because of the

design complexity of the proposed chiral patterns and the enhancement mechanism being linked only to the localized mode, it is hard to tailor the chiral resonance by simply altering geometric parameters^[41]. Although some pioneer work^[35, 36] on chiral metasurfaces linked the collective chiral responses to the propagating modes, there is still lack of clear demonstration. Consequently, the chiral responses shown in previous works were mostly spectrally fixed, which severely limits practical applications^[5, 22].

In this communication we study a novel chiral metasurface, made of an array of nanoslits milled in a ~ 100 nm thin gold layer on a sapphire substrate, that exhibits pronounced chiral optical resonant responses from visible to near IR frequencies which, thanks to the effect of surface lattice resonances^[42, 43] introduced by a propagating mode, are finely controllable by the tuning of both the lattice period and the length of nanoslits. Our results clearly highlight the impact of the propagating surface plasmon modes in controlling chiral responses in metamaterials and metasurfaces and therefore, provide a new strategy for designing chiral platforms with tailored responses as needed. Furthermore, when compared to chiral patterns made of isolated metallic nanoparticles, our metamaterial, etched in a continuous metal film, allows electrical conductivity across the film, thus enabling applications for optoelectric devices as well.

The proposed chiral pattern, a square lattice metamaterial array with unit cell period p , is illustrated in Figure 1. Each metamolecule is made of 4 nanoslits of identical length a and width b separated by a gap g , in a chiral arrangement where each slit has a 90 degree rotation with respect to the preceding one and is aligned with its long axis to the center of the following one, in either clockwise or anticlockwise rotation, to determine the handedness of the isotropic chiral pattern. Figures 1b and 1c show scanning electron

microscope images over a magnified area of a $40 \times 40 \mu\text{m}^2$ metamaterial array (50×50 unit cells) for the same sample design with opposite handedness ($a=350 \text{ nm}$, $b=100 \text{ nm}$, $g=50 \text{ nm}$, $p=810 \text{ nm}$), fabricated by focus ion beam (FIB) milling of the gold film, while arrays with the same unit cell size and different lattice constant are also prepared.

Based on the resonant profile of a single nano-slit, the chiral configuration will exhibit overlapping of E-field and H-field hot spots occurring around the 4 gaps^[6, 39], which means that the localized plasmon mode given by the coupling between adjacent nano-slits in one unit cell contributes to the enhancement of optical chiral response. In order to investigate the nature of the plasmon mode we perform full-wave numerical simulations for the left-handed pattern using the same geometrical parameters specified in Figure 1 for the fabricated metamaterial and report the results in Figure 2. The metamaterial shows a pronounced peak in the Circular Dichroism (CD) spectra around 720 nm , which is illustrated by the red dash curve in the figure 2a for the simulated structure and matches well the experimental results displayed in the figure 2b. The chiral metasurface was measured under normal illumination by a micro-spectrophotometer (Jasco MSV-5200) coupled with broadband quarter waveplates (Thorlabs) for generating circular polarized light (details in supporting information). It can be noticed that the wavelength of the CD peak is approximately two-fold the length of the nanoslits, which matches the fundamental plasmon resonance of nanoslits^[6, 44]. From this it can be concluded that the chiral nature that stems from the coupling among the nanoslits in one unit cell is predominantly based on the excitation state of the single nanoslits. In order to gain a better insight, we map the electric field distribution at resonance (720 nm) between the substrate and the gold layer in figures 2c and 2d corresponding to excitations by left

(LCP) and right (RCP) circular polarized light, respectively: the distributions of the hot spots related to the enhancement of the electric field show a clear dependence on the polarization of the incident light, resulting in dramatic asymmetric transmittance of RCP and LCP.

To confirm the chiral nature of the proposed metasurface, we carry out Circular Dichroism (CD) measurements for both enantiomers. Figures 3a and 3b show transmittance spectra for left and right enantiomers respectively, where clear differences can be observed in the transmittance spectra of LCP (blue curve) and RCP (red curve) together with a complementary behavior for the two handedness. The CD spectra, which can be retrieved by the transmittance by using the formula $\theta \text{ (deg)} = 33(\log(T_{LCP}) - \log(T_{RCP}))$, are reported in Figure 3c for both enantiomers: they are almost perfect mirror images and exhibits a huge ellipticity of nearly 4 degrees at 703 nm which is comparable with the chirality strength given by more elaborate three dimensional structures that require far more complex fabrication techniques. The FWHM of the resonance in the CD spectra is about 36 nm which could be a significant superiority for this chiral metasurface to become a promising candidate for chiral-molecule sensors. Because the mechanism is that different molecular chiral structures usually show differences in CD spectral shifts that is, however, very slight when we use chiral metasurface to sense them^[5], thus, a sharper peak benefits to detect subtler differences in spectral shifts in order to improve the sensor's sensitivity.

Beside the pursuit of pronounced optical chiral response in the visible range, tunability, to a great extent, manifests the practicality, thus is becoming a more and more important and desirable feature, which so far has been achieved by varying either the

chiral layer thickness^[45] or the refractive index of the adjacent dielectric environment^[46]. Here we propose a much more straightforward strategy to achieve tunable chirality within the same chip: since the resonance of the nanoslit is ultimately responsible for the overall resonance of the chiral arrangement in the metamolecule, on the one hand we can shift the CD peak by simply tuning the length of nanoslits, and on the other hand we can vary the lattice period to modify the cell-cell interaction via their the near-field interference, which is ascribed to the propagating mode. To verify this, we prepared three arrays with identical unit cell ($a=350$ nm, $b=100$ nm, $g=50$ nm) but various lattice periods ($p=810$ nm, 830 nm, 850 nm) and perform both numerical simulations and experimental measurement of the spectra which are reported in Figure 4a and 4b and show a good agreement. As expected we observe red shifting of the CD resonance when the lattice period increases from 810 nm to 850 nm. The CD spectral shift is proportional to the increment in the period (in experiments, ~ 7 nm shift for a period increase of ~ 20 nm), which indicates that, beside the intrinsic chiral responses generated by the localized plasmon mode, the lattice period plays a role by modifying the cell-cell coupling. This proportional tunability is extremely important because it gives it enables the superfine control of the chiral response which means, for example, that a device can be designed to exactly match the resonance of multiple targets.

The underlying mechanism can be understood as follows: the change in the lattice period affects the interference of the propagating plasmon mode across unit cells, which modifies the collective CD spectra. Intuitively, the chiral metasurface can be decomposed into 4 groups of gratings supporting propagating modes (Fig. 4c) that, when combined together, form a chiral shape resulting in a chiral asymmetry of the propagating plasmon

mode. For a clear visualization, we simulate an infinite array and plot, for a single unit cell, the field map distributions at resonance ($\lambda = 735$ nm for $p = 830$ nm) of the electric field component perpendicular to the film under circularly polarized illumination at both gold-sapphire interface (figures 4d and 4e). When comparing the Figures 4d and 4e, which show the surface plasmon resonance under the excitations of LCP and RCP waves, an asymmetric propagating feature appears in the near-field, which enables circular polarization dependent coupling between neighboring chiral unit cells and results into a collective nature of the CD. Therefore, through tuning the lattice period, the coupling is modified effectively to make the chiral response controllable and tunable. It has also to be noted that the magnitude of the CD peaks drops with the increase of the lattice period. This phenomenon can be heuristically explained by considering the combination of the localized mode and the propagating mode: the unit cell dimension and geometry imposes the localized mode while there is an optimal value for the lattice period to best match the propagating mode to the localized one, which results in the strongest chiral response. As a reference, we have also fabricated the inverse chiral patterns, in which nanorods replace the nanoslits with identical parameters for the unit cell and lattices periods ($p=810$ nm, 830 nm, 850 nm): in this cases we do not detect any obvious shift in the CD (details in supporting information), which further indicates that the cell-to-cell coupling results from the propagating mode on the metallic surface and it not present in the inverse structures.

While the adjustment of the lattice period is perfectly suited for fine tuning of the resonant chiral response of the metasurface, it cannot be used to achieve large tuning ranges, which can instead be obtained by engaging the resonant feature of the localized plasmon mode simply by changing the length of the nanoslits. We experimentally

demonstrate this ‘coarse’ tuning strategy by fabricating samples with nanoslit lengths of 300 nm, 350 nm and 400 nm: here the change in the length of the nanoslits will induce a localized plasmon resonance shift for the single slit and, consequently, for the unit cell. Figures 5a, 5b and 5c show the transmittance spectra of three left-enantiomer samples with different nanoslits length for both LCP (blue curve) and RCP (red curve). The increase in the slits length produces, as expected, a red shift of the optical chiral response across a significant wavelength range (>100 nm) which can be easily appreciated in Figure 5e where CD spectra for the three samples are shown together and can be easily compared. Therefore, the interplay between localized and propagating plasmon modes allows not only having the chiral response of our metasurface to span a broad tunable range, but also doing so in fine steps, which could be summarized as having a ‘coarse tuning + fine tuning’ knobs corresponding to the modification of the localized and propagating mode, respectively.

In summary, we have proposed and experimentally demonstrated a tunable chiral metasurface showing strong (~ 4 degrees of CD) and narrow linewidth (~ 36 nm) intrinsic chiral optical response, which can be finely controlled over a broad range in the visible spectrum. The remarkably narrow linewidth of the CD peak could highly benefit various potential applications, such as chiral bio-molecule sensing. Our strategy highlights for the first time the coupling between the localized and propagating modes, thus providing an extra degree of freedom in the design of optimal chiral metasurfaces, and effectively providing two independent ‘knobs’ to achieve large tuning intervals with fine adjustment within them.

Experimental Section

Fabrication: The chiral metasurface is fabricated by a two-step process: first, the cleaned sapphire substrate is coated with 100 nm Au layer by thermal evaporation and then, a focus ion beam writer (FEI Helios 650) operated at 30 kV and 7.7 pA was used to generate the chiral pattern.

Experimental Measurements: One home-made sample holder into which quarter waveplate can be inserted is employed in this system. We keep the fast axis in horizontal as shown in Figure S1 (details in supporting information) and set the incident light with linear polarization, while at 45 or -45 degrees, relative to the fast axis direction. Respectively, we have left and right circular polarized light generated after the quarter waveplate and then going through the sample. The baseline was made by measuring the clean sapphire substrate without chiral structures.

Numerical Simulation: All the simulation work was completed by the software FDTD Solutions. The reflective index of sapphire substrate (Al_2O_3) and Au film was from Palik based on the built-in database of the software. In the X and Y directions, we use Bloch boundary condition because of the periodic property of the metasurface, while for the Z direction, perfectly matched layer (PML) boundary was employed. The mesh size of (5 nm, 5 nm, 5 nm) was applied for the structure of metasurface.

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Figures

Figure 1. (a) Schematic illustration of the chiral metasurface. (b) SEM images of left-handed chiral metasurface. (c) SEM images of right-handed chiral metasurface.

Figure 2. (a) Simulated and (b) experimental CD spectra for the left-handed chiral metasurface and electric field intensity maps of the metasurface under (c) left and (d) right circularly polarized illumination, at the resonant wavelength (720 nm).

Figure 3. The mirror image is obtained experimentally by the CD spectra of the left and right handed chiral metasurface. (a) The absolute transmittance spectra corresponds to left (blue curve) and right (red curve) circular polarized illumination of the left-handed and (b) right-handed enantiomer metasurface. (c) The CD spectra transferred from the transmitted spectra of the both handed metasurface, which form a perfect mirror image.

Figure 4. Controlling the chiral response by varying the lattice period. (a) Simulated CD spectra and (b) experimental differential transmittance spectra for the array with identical unit cells but various periods of 810nm (black curve), 830nm (red curve) and 850nm (blue curve). (c) The chiral metasurface is decomposed into 4 groups of gratings labeled with different colors. For the array with the period of 830nm, the electric field Z-component distributions excited by LCP and RCP on the (d, e) bottom surface.

Figure 5. Controlling the chiral response by modifying the localized mode. The absolute transmittance spectra of chiral metasurface with the various nanoslits length from (a) 300nm ($a=300\text{nm}$, $b=50\text{nm}$, $p=750\text{nm}$) to longer nanoslits of (b) 350nm ($a=350\text{nm}$, $b=50\text{nm}$, $p=810\text{nm}$) and (c) 400nm ($a=400\text{nm}$, $b=50\text{nm}$, $p=890\text{nm}$) under the illumination of LCP (blue curve) and RCP (red curve) respectively. (d) SEM images of the chiral metasurface with the various nanoslits length. (e) The CD spectra of the chiral metasurface with various length of nanoslits, in which the obvious blue shift can be observed corresponding to the shortening of the nanoslit.

The table of contents entry: The enhanced chiral response exhibited by chiral metamaterials and metasurfaces is generally attributed to the effect of localized surface plasmon resonances, which are mostly determined by the constituent materials and structure geometry: how the presence of propagating surface plasmon modes influences chirality is still short of investigation. Herein, we introduce a novel chiral metasurface composed of a chiral arrangement of nanoslits carved in a continuous metal film (inset left), in which propagating surface plasmon modes are proved to be powerful tools to control the metasurface's chiral response. Numerical simulations help us devise a straightforward approach to the metasurface's design and through which, by modifying the coupling between localized and propagating modes, we experimentally achieved controllable and remarkably strong chiral responses (inset right). Our finding provides an extra degree of freedom in manipulating chiral responses, which may aid in the further design of chiral plasmonic structures.

Keywords: *Controllable, tunable, chirality, metasurface, continuous plasmonic structure*

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