Conditions for enhancing chiral nanophotonics near achiral nanoparticles

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Conditions for Enhancing Chiral Nanophotonics near Achiral Nanoparticles

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Supporting Information

ABSTRACT: The interaction of circularly polarized light with matter is the basis for molecular circular dichroism spectroscopy, optical spin manipulation, and optical torques. However, chiroptical effects are usually hampered by weak chiral light–matter interaction. Nanophotonic structures can enhance optical intensity to boost interactions, but magnifying chiral effects requires that the near field remains chiral in the process. Here, we propose the conditions and limits for enhancing different chiroptical effects near achiral metasurfaces with maximum chirality of the evanescent fields. We illustrate these conditions with arrays of metal and dielectric nanodisks and decompose their distinct electromagnetic metrics into propagating and evanescent fields. We prove that a nanostructure cannot be universally optimal for different chirality metrics and therefore applications. For example, arrays tailored for enhanced spin excitation with spatially uniform circular polarization destroy circular dichroism. Conversely, we predict a limit of maximum attainable circular dichroism in highly evanescent Fourier orders. We illustrate these conditions in diverse chiroptical applications.

KEYWORDS: Optical chirality, Near field, Evanescent waves, Plasmonic resonances, Mie resonances, Nanoparticle arrays

Chiral molecules occur in two forms, mirror images of each other, known as enantiomers. They are essential in chemistry, biology, and medicine. The distinct interactions of enantiomers with the human body can manifest harmlessly (e.g., as a difference in odor) or drastically (e.g., as side effects of a drug). Light has two different helicities (right or left circular polarization) to probe chiral molecules, providing conformational information down to the nanometer scale. Circular dichroism (CD), the differential absorption of left and right circularly polarized light by chiral molecules, can be used to distinguish between enantiomers or to create an excess of a specific enantiomer. Circularly polarized light can also interact selectively with other useful ways. It contains angular momentum, which can rotate objects via optical torques for nanomanipulation. It can initialize and read out electronic and nuclear spins in semiconductors and switch magnetization optically. In monolayers of transition metal dichalcogenides (TMDs), chiral light allows the population of specific spins in momentum valleys. Thus, chirality is also a resource for information technology.

The main impediment to the sensitivity of chiral light–matter interactions is often weak contrast. The difference in absorption between opposite helicities of light in CD is usually 3–4 orders of magnitude weaker than the value of absorption itself. Nanophotonic near-field enhancement is being explored to enhance chiral light–matter interactions, with the focus mostly on CD using dielectric and metal nanostructures or films. Inherently chiral nanophotonic systems designed to enhance CD induce an undesired background over the CD signal from the analyzed molecules. Two considerations make the design of general nanophotonic systems for the enhancement of chiral light–matter interactions challenging. First, apart from enhancing intensity in the near field, the nanostructures must also preserve the incident circular polarization to retain chiral contrast without adding an intrinsic bias. For most applications, it is desirable to have an identical response for both circular polarizations of light by using an achiral nanostructure. Second, different chiral light–matter interactions depend on different metrics of the electromagnetic field, requiring the design of separate nanostructures tailored to various effects. In other words, the same nanostructure might not be universally optimal for all chiroptical effects.

Here, we analyze the conditions for the nanophotonic enhancement of chiral light–matter interactions, focusing on enhancing the near-field intensity while preserving circular polarization. We consider arrays of achiral nanodisks as an illustrative example and study the enhancement of various chiral interactions over an extended area near the array. We
first discuss the different metrics characterizing chiral light–matter interactions. We numerically demonstrate that the resonant near-field enhancement in metallic nanodisk arrays is accompanied by low CD and a detrimental spatial variation of the handedness. We then introduce a Fourier decomposition of the chirality metrics into propagating and evanescent orders to explain these results. Based on the Fourier decomposition, we analytically derive the limits on the simultaneous enhancement of different chirality metrics and formulate the conditions to maximize them in evanescent fields. We show that dielectric nanodisk arrays can enhance near-field chirality within these limits. Although exemplified with nanodisk arrays, the conditions of enhancement that we derive apply to general nanophotonic systems and will help to design tailored nanostructures to boost various chiral effects.

**METRICS FOR CHIRAL LIGHT–MATTER INTERACTIONS**

The polarization of a plane wave is usually expressed in terms of Stokes parameters. The relevant parameters for quantifying circular polarization are the Stokes parameters $I$ (the total electric field intensity) and $V$ (the difference between the intensities of left and right circularly polarized waves). The degree of circular polarization (DOCP) of the wave is defined as $\text{DOCP} = V/I$. Purely circularly polarized waves have DOCP $= \pm 1$. For plane waves, all chiral light–matter interactions depend on the Stokes parameters. Close to a nanoparticle, however, several propagating and evanescent waves contribute to the fields. Various methods have been proposed to extend the theory of polarization to the near field.25,26 Here, we focus on the chirality metrics in the near field which affect prominent chiral light–matter interactions.

CD typically involves the electric and magnetic dipole moments of the molecule1 and hence depends on both the electric and magnetic fields through Lipkin’s chirality parameter $C = \frac{1}{\omega^2} \text{Im} [\mathbf{E} \cdot \mathbf{B}]$.27,28 Lipkin’s chirality in the near field is related to the Stokes parameter $V$ in the far field through the chirality flux.29,30 The differential absorption of circular polarizations is proportional to $G^* C$, where $G^*$ is the imaginary part of the mixed electric–magnetic dipole polarizability of the molecule. In the limit of low CD, the total absorption for both circular polarizations is proportional to the electric energy density $U_e = \frac{\omega^2}{\epsilon_0} |\mathbf{E}|^2$. CD enhancement can be quantified by the dissymmetry factor $g$, which is the ratio of the differential absorption to the average absorption of the two polarizations. It is thus proportional to the ratio of Lipkin’s chirality and the electric energy density, $g \propto \frac{C}{U_e}$. The mathematical forms of the Stokes parameters and CD enhancement are similar: $I$ and $U_e$ are metrics of total intensity, whereas $V$, DOCP, $C$ and CD enhancement are metrics of chirality quantifying the difference between opposite circular polarizations.

Other chiral light–matter interactions do not depend directly on the magnetic field, as the magnetic dipolar response of the material is negligible at optical frequencies. Examples include optical torques and spin polarization, which usually involve only electric dipoles. The electric field vector at a given point rotates in a plane and traces an ellipse (solid ellipse in Figure 1a).31 Therefore, we define the Stokes parameters and the DOCP locally on this plane of polarization and refer to them as $I_{\text{2D}}, V_{\text{2D}},$ and $\text{DOCP}_{\text{2D}}$. Expressing $V_{\text{2D}}$ as a vector perpendicular to the plane of the ellipse gives the electric spin density.32 The optical torque on a small absorbing particle is proportional to the electric spin density.33 $V_{\text{2D}}$ is also the relevant electromagnetic field metric for generating optical magnetization using the inverse Faraday effect.34-36

Although the local polarization ellipse in the near field lies in an arbitrary plane, its projection on a fixed plane becomes relevant in planar structures such as substrates, thin films like 2D materials, metasurfaces, or arrays. We can then define the Stokes parameters in terms of the in-plane projection of the electric field (dashed ellipse in Figure 1a) and denote the projected parameters by $I_{\text{2D}}, V_{\text{2D}},$ and $\text{DOCP}_{\text{2D}}$. As an example, a small disk constrained to rotate on the plane would experience an optical torque proportional to $V_{\text{2D}}$. For transition metal dichalcogenide (TMD) monolayers, optical absorption is proportional to $I_{\text{2D}}$, and the differential spin-valley excitation to $V_{\text{2D}}$.37 Exciting only a given spin or valley requires therefore high DOCP$_{\text{2D}}$.

Figure 1. A metallic nanodisk array provides strong intensity enhancement but poor chirality. (a) A silver nanodisk array with radius $R = 75 \text{ nm}$, height $H = 40 \text{ nm}$, and period $P = 400 \text{ nm}$ is illuminated from above by a left circularly polarized plane wave. The near field is evaluated on a plane $h = 10 \text{ nm}$ above its surface. The local three-dimensional electric field $E_{2D}$ traces an ellipse with time (solid) and its projection on the plane (dashed) gives the in-plane field $E_{2D}$. (b) Reflectance from the array (black) and enhancement of the chirality metrics at a point above the edge (red point). Maps of the enhancement of (c) the intensity metrics and (d) the chirality metrics at $\lambda = 600 \text{ nm}$ in the unit cell plane above the disk (outlined by dashed circles). Only half of the unit cell is depicted in each map due to symmetry.
Figure 2. Fourier decomposition provides insight into the chiral fields near metallic nanodisks. (a) Schematic of Fourier decomposition depicting the in-plane wavevectors (dashed arrows) and the vertical variation in field intensity (pink) for the lowest Fourier orders. (b) Fourier decomposition of the intensity metrics $U_e$ (left) and $I_{2D}$ (right) at $\lambda = 600$ nm for the metallic nanodisk array in Figure 1, on the plane 10 nm above the surface of the array. (c) Fourier decomposition of the chirality metrics: circular dichroism (left) and degree of circular polarization (right).

### CHIRALITY NEAR AN ARRAY OF METALLIC NANODISks

We first demonstrate that, at resonance, an array of silver nanodisks enhances the near field intensity but shows poor chirality metrics. A left circularly polarized plane wave illuminates the nanodisks from above at normal incidence (Figure 1a). We arrange the nanodisks in an array to obtain field enhancement over an extended region and evaluate the near field on a plane above the nanodisks.

The reflectance of the array exhibits a localized plasmon resonance peaking near the wavelength $\lambda = 600$ nm (Figure 1b, black line). The Stokes parameter $V_{2D}$ (purple line) at a point near the edge of the nanodisk (red dot in the schematic) also shows resonant behavior but with a negative value. The sign reversal indicates that the electric field near the nanodisk rotates in a sense opposite to that of the incident light. The enhancement of Lipkin’s chirality $C$ is negligible in comparison.

The near-field intensity is the strongest near the edge of the metallic nanodisk at resonance (Figure 1c). The electric energy density $U_e$ (left) is enhanced by more than 50 times compared to a plane wave. The enhancement in the Stokes parameter $I_{2D}$ (right) is lower because of the significant out-of-plane electric field. In contrast, the chirality metrics of circular dichroism (CD enhancement) and degree of circular polarization (DOCP2D) are not preserved (Figure 1d). CD enhancement, which is the ratio of $C$ and $U_e$, is saturated $\pm 1$ for circularly polarized plane waves, is suppressed everywhere (left) due to a higher enhancement of $U_e$ compared to $C$. The situation is different for DOCP2D = $V_{2D}/I_{2D}$ (right). At the center of the nanodisk, DOCP2D is unity, as required by symmetry. However, as we approach the edge of the nanodisk, DOCP2D changes abruptly from positive to negative. DOCP2D recovers the value of unity at the corners of the unit cell due to symmetry. The existence of regions with opposite signs leads to suppression of DOCP2D when averaged over the unit cell. We conclude that the low CD and the cancellation in DOCP2D make the metallic nanodisk a poor choice for enhancing chiral interactions over large areas.

### FOURIER DECOMPOSITION

We perform a Fourier decomposition of the near field including both propagating and evanescent waves (Figure 2a) to understand the poor chirality metrics of metallic nanodisks. If $\hat{F}(\mathbf{r})$ is a field amplitude vector such as $\hat{E}$ or $\hat{B}$, we have the Fourier decomposition $\hat{F}(\mathbf{r}) = \sum_{mn} \hat{F}_{mn} \exp[i\mathbf{k}_{mn} \cdot \mathbf{r}]$, where $\mathbf{r} = (x, y)$ and $\mathbf{k}_{mn} = \frac{2\pi}{\lambda} (m\hat{x} + n\hat{y})$ is the in-plane wavevector. The Fourier coefficients $\hat{F}_{mn}$ are computed by integrating the fields over the unit cell

$$\hat{F}_{mn} = \frac{1}{p^2} \int \hat{F}(\mathbf{r}) \exp[-i\mathbf{k}_{mn} \cdot \mathbf{r}] dS$$

(1)

The incident wave and the reflected wave combine to form the $(0, 0)$ order with a standing wave intensity pattern in the vertical direction (pink). All the higher Fourier orders are evanescent because the lattice spacing is smaller than the wavelength in the background medium. Their in-plane wavevectors are shown with dashed arrows. The Fourier amplitudes of these evanescent orders decay vertically with a decay constant $k_{mn,z}$ given by the dispersion relation $k_{mn,z}^2 = \left| \mathbf{k}_{mn} \right|^2 - k_0^2$ where $k_0$ is the wavenumber in the background medium. As the Fourier order increases, so does the in-plane wavevector accompanied by a quicker evanescent decay.

Adding up the contributions from all the Fourier orders yields the average values of the intensity and chirality metrics over the unit cell (see Methods for details). For a metric $M (U_e, C, I$ or $V)$, the average value on a plane in the unit cell $(M) = \sum_{mn} M_{mn}$ where $M_{mn}$ are the Fourier coefficients of the metric. Both the CD enhancement and DOCP2D of an evanescent Fourier order remain invariant with distance away from the nanodisks (Supporting Information section S5).

The lowest evanescent orders $(0, \pm 1)$ and $(\pm 1, 0)$ dominate the intensity metrics above the silver nanodisks (Figure 2b). Almost all the energy density is confined to the $3 \times 3$ Fourier orders surrounding $(0, 0)$. The same is true for the Stokes parameter $I_{2D}$, although higher Fourier orders have lower values of $I_{2D}$ than $U_e$ due to their vertical field components. The chirality metrics of the various Fourier orders are significantly different (Figure 2c). The CD enhancement is positive but lower than unity for the $(0, 0)$ order. Indeed, the CD enhancement can vary along the direction of propagation for standing waves, attaining values both below and above that of circularly polarized plane waves.\cite{28,36} Meanwhile, the CD enhancement is negligible for all the higher orders. On the other hand, circular polarization is perfect for the $(0, 0)$ order, with DOCP2D = 1 as required by symmetry. The higher orders $(0, \pm 1), (\pm 1, 0)$, and $(\pm 1, \pm 1)$ are highly circularly polarized, but with opposite handedness to the incident wave with DOCP2D values close to $-1$. The negative values of DOCP2D in the evanescent orders combined...
with the positive value for the (0,0) order result in spatial cancellation of DOCP2D.

Evanescent waves have a general limitation: high in-plane circular polarization DOCP2D always results in low CD. As we prove in Supporting Information section S1, for an evanescent Fourier order, Lipkin’s chirality is given by

$$C_{mn} = \frac{k_{mn,x}}{k_{mn,z}} \Re \left( \tilde{E}_{mn,x} \tilde{E}_{mn,y}^* \right)$$

where $\tilde{E}_{mn,x}$ and $\tilde{E}_{mn,y}$ are the in-plane components of the Fourier coefficient of the electric field. When the in-plane circular polarization is perfect (DOCP2D = ± 1), $\tilde{E}_{mn,x}$ and $\tilde{E}_{mn,y}$ are 90° out of phase. As a result, $\Re \left( \tilde{E}_{mn,x} \tilde{E}_{mn,y}^* \right) = 0$, leading to Lipkin’s chirality and CD enhancement being zero. Thus, the chirality metrics DOCP2D and CD are incompatible in the near field, in complete contrast to propagating plane waves in the far field where they go hand-in-hand. The incompatibility of the chirality metrics makes it impossible to design nanostructures exploiting evanescent waves to enhance near-field intensity while preserving both CD enhancement and DOCP2D simultaneously on a plane.

If we relax either condition, of intensity enhancement or circular polarization preservation, it is indeed possible to obtain CD enhancement, even above unity. It is known that superchiral fields can be created in standing waves with intensity cancellation.28 We prove in Supporting Information section S2 that a loss of in-plane circular polarization permits CD enhancement, only preserved. Obtaining significant CD enhancement requires exciting high in-plane wavevectors and having an optimum polarization to match eq 2.

### CHIRALITY NEAR AN ARRAY OF SILICON NANODIKS

The limiting factor for chirality near metallic nanodisks is the spatially varying sign of the DOCP2D (Figure 1d), which is due to the boundary conditions for the electric field at a metal-dielectric interface (Supporting Information section S3). The primary culprit is the negative permittivity of the metallic nanodisk. Replacing it with a dielectric nanodisk of positive permittivity can get rid of sign cancellation. High-index dielectric nanoparticles have been suggested for CD enhancement29 and chiral emission37 because they can support electric and magnetic dipoles simultaneously. Silicon metasurface designs have been proposed based on this idea.30,11

Next, we show how silicon nanodisks provide strong intensity enhancement with a homogeneous circular polarization without sign cancellation. We use silicon nanodisks in a square array (Figure 3a). We evaluate the fields in a plane below the surface because retardation due to the high refractive index of silicon results in a strong front-back asymmetry with stronger fields below the disk (Supporting Information Figure S1). The nanodisk array has two reflection resonances (Figure 3b), arising from magnetic and electric dipole modes...
for chirality enhancement. We focus on the magnetic dipole resonances makes silicon nanodisks a good candidate for excitation of spectrally overlapping electric and magnetic intensity is in the wavelength where transmittance is minimized. Most of the transmission side of the structure and we operate at a central (0, 0) order because the observation plane is on the edge of the disk, almost vanishing outside (Figure 3c).

In the Fourier decomposition, there is negligible intensity in (3d), except at a small area near the edge where the intensity is low. All the Fourier orders are almost perfectly circularly polarized. At this wavelength, however, the silicon disk does not provide enhanced CD, which is low everywhere on the unit cell and particularly in regions of high intensity. The reason is that all the energy is concentrated in the almost perfectly circularly polarized evanescent orders. As demonstrated above, such evanescent orders necessarily have low CD. The (0, 0) order, which is a propagating order that remains circularly polarized due to symmetry, has a CD enhancement of unity but very low intensity.

INCOMPATIBILITY AND LIMITS OF CHIRALITY METRICS

Finally, we demonstrate that enhanced circular dichroism is achievable over a large area near silicon nanodisk arrays, but only by compromising on the degree of circular polarization. We calculate the average values of the chirality metrics on a plane 10 nm below the nanodisk (see Methods) for different periods of the array. At a given wavelength, the silicon nanodisk array can offer high VCD and DOCP2D, simultaneously (Figure 4a). The points marked with a circle in Figure 4 correspond to the maps in Figure 3c,d. Although there is a large enhancement in VCD with almost perfectly preserved DOCP2D at the marked point, it produces poor CD (Figure 4b). For all periodicities, wavelengths with high DOCP2D have low CD, and vice versa (Figure 4c). For a combination of geometry and wavelength where C and CD are simultaneously enhanced (marked with a triangle), VCD is low because of the spatially varying sign of DOCP (Figure 4d). The Fourier decomposition of the metrics indicates that high CD arises from the evanescent orders (0, ±1), (±1, 0), and (±1, ±1) with almost linear in-plane polarization.

CD enhancement greater than unity occurs for the lowest period (P = 320 nm) where the evanescent Fourier orders have the highest in-plane wavevectors (Figure 5, blue circles), consistent with our finding that high in-plane wavevectors provide maximum CD enhancement. CD enhancement is only slightly above unity because only the lowest evanescent orders are excited. If we consider silicon nanodisk arrays of various geometries, they support evanescent orders spanning a range of CD enhancement values (Figure 5, dark blue dots), all under the maximum limit of eq 3 (yellow line). Some of the orders approach the theoretical maximum limit and have CD > 1. However, enhancing the average CD over the nanodisk array using high-CD evanescent orders requires that they have higher intensity than other low-CD orders. Further structure design to suppress lower evanescent orders while exciting higher orders with the optimal polarization is thus a promising path to obtain high CD enhancement together with intensity enhancement over large areas. Our Fourier decomposition method can be extended to individual nanostructures by using an angular spectrum representation with continuous evan-
escent wavevectors. Our results also apply to situations including substrates and molecules dispersed in a thin film above the nanostructures (Supporting Information sections S5 and S6).

## CONCLUSIONS

We have presented conditions for the nanophotonic enhancement of chiral light–matter interactions based on a Fourier decomposition of the near field. Preserving circular polarization and enhancing circular dichroism are mutually contradictory if the intensity is enhanced by evanescent waves on a plane parallel to the nanostructures. Consequently, tailoring metasurfaces for optimal molecular circular dichroism is incompatible with good performance for other chiroptical applications such as spin-selective excitation in semiconductors or optical torque. Silicon nanodisk arrays offer a flexible platform for enhancing different chiroptical effects. By tuning geometry and wavelength, they can provide near-field intensity enhancement optimized for either circular polarization or circular dichroism. To conclude, we propose to achieve large-area circular dichroism above unity on a metasurface dominated by highly evanescent orders. Our results define the limits on the ability of nanophotonic platforms to enhance different chiral light–matter interactions simultaneously and provide design rules for individual applications.

## METHODS

### Numerical Simulation.

We perform the numerical simulations using the surface integral equation (SIE) method for homogeneous and periodic structures. The edges of the nanodisks are rounded with $r = 20$ nm. The nanodisks are placed in a homogeneous background permittivity $\varepsilon_r = 1.5$ corresponding to the geometric mean of the permittivities of air and glass. The material parameters are taken from Johnson and Christy for silver and Green for silicon.

## Averaging Chirality Metrics.

The physically meaningful way to evaluate the average values of the chirality metrics is as follows. For the metrics $U_I$, $I_{2D}$, $C$, and $V_{2D}$, the averages can be found by direct spatial averaging $\langle x \rangle = \frac{1}{P} \int x \, dS$, where $x$ is one of the four metrics. However, DOCP$_{2D}$ and CD enhancement need to be evaluated as the ratios of the averages of the other metrics: $\langle \text{DOCP}_{2D} \rangle = \frac{\langle \text{DOCP}\rangle}{\langle I \rangle}$ and $\langle \text{CD} \rangle = \frac{\langle \text{C} \rangle}{\langle I \rangle}$.

The average values of the intensity and chirality metrics in the unit cell are the sums of their respective values for the Fourier orders. For a function of the form $\mathbf{G}^* \cdot \mathbf{F}$, where $\mathbf{F}$ and $\mathbf{G}$ are fields (E or B) with Fourier coefficients $\mathbf{F}_{mn}$ and $\mathbf{G}_{mn}$ given by eq 1, the spatial average in the unit cell is

$$\langle \mathbf{G}^* \cdot \mathbf{F} \rangle = \frac{1}{P^2} \int \mathbf{G}^*(\mathbf{r}) \cdot \mathbf{F}(\mathbf{r}) \, dS = \sum_{mn} \mathbf{G}^*_{mn} \cdot \mathbf{F}_{mn}$$

which is a form of the Parseval relation. The spatial averages can be found by summing the corresponding values of the Fourier orders.

$$\langle M \rangle = \sum_{m,n} M_{mn}$$

where $M$ is a metric and $M_{mn}$ are Fourier coefficients of the metric evaluated from the coefficients of the respective fields.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphotonics.9b01200.

- Impossibility of simultaneous preservation of DOCP and CD; limits of CD enhancement in evanescent Fourier orders; boundary conditions and retardation; multipole decomposition; decay of chirality metrics away from the nanodisks; influence of substrate on chirality metrics (PDF)

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### Notes

The authors declare no competing financial interest.

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