Giant Enhancement of the Chiro-Optic Response of Chiral Plasmonic Media Using Structured Light

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ABSTRACT

We propose an approach to achieve a great enhancement and broad spectral tunability of the chiro-optic response of plasmonic metasurfaces by exploiting their interaction with light of complex polarization, endowed with both spin (SAM) and orbital angular momentum (OAM). As a proof of principle, we employ full-wave computational analysis to investigate the response of a circularly polarized transverse Laguerre-Gaussian beam having a structured wavefront, incident on a chiral metasurface composed of a two-dimensional array of gold nanohelix meta-atoms. For the first time, our analysis reveals unprecedented amplification of circular dichroism, which is a manifestation of the total angular momentum of incident light being enantio-selectively transferred to the chiral medium.

Keywords: structured light, chiral metasurfaces, plasmonics, circular dichroism, biosensing.

1 INTRODUCTION

In the past decade, structured light has generated huge interest due its peculiar applications in the field of telecommunications [1-3]. As we know, cirularly polarized light or light with spin angular momentum (SAM) has a circularly rotating electric field vector along the line of propagation, while in structured light or light with orbital angular momentum (OAM), the whole wavefront is twisted leaving a singularity at the center [4]. SAM can have two states, right circularly polarized state (RCP) or left circularly polarized (LCP) state. OAM on the other hand (usually represented by l) can take infinite integer values. Light with OAM can be generated using several techniques like: spiral phase plates [5], pitch fork hologram [6] or by beam shaping using a metasurface pattern [7]. In this paper, we use a combination of SAM and OAM. To generate light carrying both OAM and SAM, a combination of polarization preserving axicon with a biaxial crystal [8] or spatial light modulators [9] can be used. Light with SAM and OAM when incident on custom designed metasurfaces – a subwavelength patterned two dimensional medium, can induce extraordinary optical phenomena [10]. It can further have applications in spin-controlled integrated photonic circuitry where optical analogues for gates, diodes etc. can be designed.

In this paper, we investigate the effects of strctured light when incident on chiral plasmonic media. Chiral media displays optical activity. Circular dichroism (CD) is an enantio-selective absorption technique, widely used to measure optical activity. CD can be obtained by taking the difference in the absorption of right and left circularly polarized light that is incident on chiral media. This activity is enhanced when the media is tethered/bonded with gold/silver nanoparticles. In the past, several techniques [11] were used to design these complex media. Chiral plasmonic media enhances CD due to the plasmonic coupling [11]. For the sake of analysis, chiral plasmonic media is approximated to an array of gold nanohelices. In the following, we demonstrate the enhancement of optical activity when structured light is used in the place of regular circularly polarized light. We also demonstrate a versatile tunability of chiro-optic response by using structured light carrying both SAM and OAM as shown in Fig.1 [12, 13].

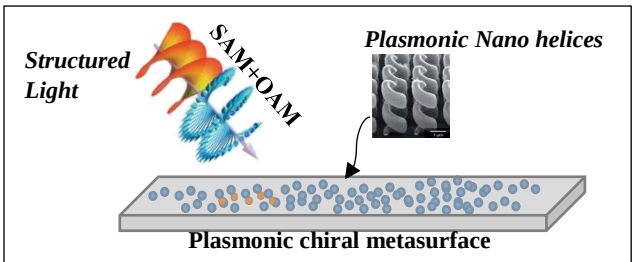

Figure 1: Pictorial representation of interaction of structured light with chiral plasmonic media

2 THEORY

The angular momentum of light can be of two types: Spin (SAM) and Orbital (OAM). By convention, SAM is represented by σ which can take values ± 1, $+1$ being RCP and -1 being LCP. OAM is represented by l and in this paper we restrict l to 0, ± 1. We used Laguerre-Gaussian (LG) beams [3] as they can carry OAM. The equation for scalar Laguerre-Gaussian beam in cylindrical co-ordinates (r, ϕ, z) reads:

$$LG_n^l(r,\phi,z) = \sqrt{\frac{2n!}{\pi(n+|l|)!}}\frac{w_0}{w_z}\left(\frac{\sqrt{2}r}{w_z}\right)^{|l|} L_n^{|l|}\left(\frac{2r^2}{w_z^2}\right)\exp\left[-\frac{r^2}{{w_z}^2} - i\left(l\phi + \frac{kr^2}{2R_z} - (2n+|l|+1)\tan^{-1}\left[\frac{2z}{kw_o^2}\right]\right)\right]$$

Here, $L_n^{|l|}$ is the generalised Laguerre polynomial of order n and degree $|l|$, while n, l are radial and OAM quantum numbers, respectively. w_z and R_z are the beam's waist and radius of curvature that are given by

$$w_z = w_0\sqrt{\left(1+\frac{4z^2}{k^2w_0^4}\right)};\ R_z = z + \frac{k^2w_0^4}{4z}$$

respectively, where w_0 is the beam waist radius at $z=0$, and $k = 2\Pi/\lambda$ is the wavenumber in the host medium. When helical plasmonic nanoparticles interact with circularly polarized light, a resonant transfer of energy from the field to the nanoparticles occurs, which depends on the photon spin coupled to the surface electron density, *i.e.* driving it in-phase or out-of-phase. Invoking a classical electron model, the resonant coupling can be understood as an in-phase generation of an electric current that is manifest by an enhanced absorption rate. In a helical structure, the electron current induces a magnetic dipole moment along with an electric dipole moment due to charge displacements and build-up at the boundaries. The mutual orientation of these two dipoles depends on the field spin and helicity of the structure and their dot product determines the CD value. There can be one or more resonances depending on the geometry of the nanohelix structure and its placement with respect to the wave vector. Furthermore, placing mutiple nanohelices in the beam path will lead to dipole-dipole coupling between these structures and as a consequence, multiple hybridized resonances. In this study, we discuss how the combination of OAM with circularly polarized light changes the spectral response of the system. Since, OAM results in a spatially distributed helical wavefront, it shows a profound effect when it is incident on helices by driving the surface charges in-phase and out-of-phase depending on the direction of twist.

3 COMPUTATIONAL MODEL

We analyze the interaction of structured light interaction with chiral plasmonic media using the electromagnetics RF module of COMSOL 5.2a (www.comsol.com). Gold nanohelices (GNH) with a major radius of 30nm and an axial pitch of 60nm are used for the analysis. The GNH are placed in a spherical domain surrounded by a shell of a perfectly matched layer (PML), which completes the compuational doamain. A scattering boundary condition is imposed on the outer boundary of the computational domain. The time harmonic E field within the region satisfies:

$$\nabla\times\left(\mu_r^{-1}\nabla\times E\right) - k_0^2\left(\varepsilon_r - j\frac{\sigma}{\omega\varepsilon_0}\right)E = 0$$

In this equation, ε_r, μ_r and σ are the relative permittivity, permeability and conductivity of the system, respectively. Here ε_r is obtained from a dielectric function [14] as a function of λ. Coming to the incident light, Laguerre-Gaussian modes $LG_0^{0,\pm1}$ (OAM (l) of 0, 1 and -1) along with SAM (σ of +1 and -1) are used. We consider the total angular momentum ($j=\sigma+l$) as we analyse the results. In the absence of OAM $(l=0)$, total angular momentum will comprise of SAM only, and can take values of ±1. With the addition of OAM, j can take values $0,\pm2$. The medium surrounding the GNH has a relative permittivity of 2.25, which is a typical value for an organic host polymer material, polyvinyl chloride (PVC). We conducted a parametric analysis by varying the incident

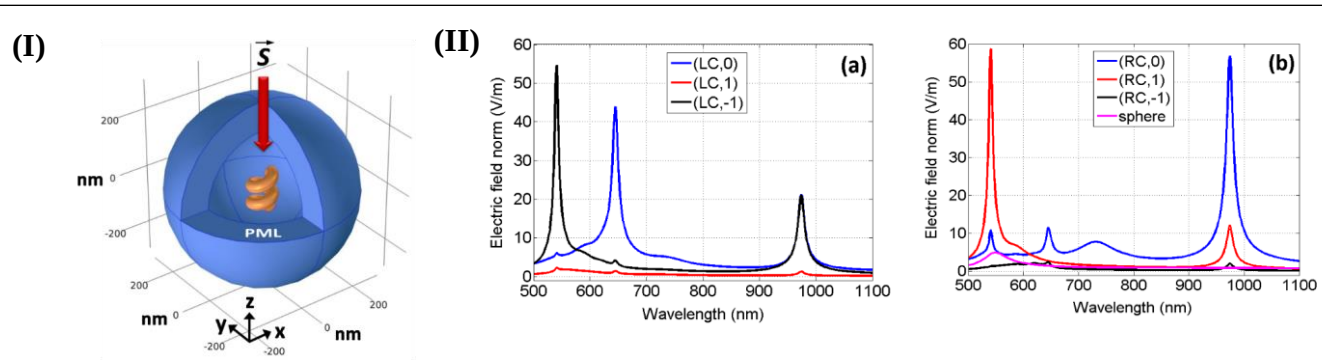

Figure 2: (I) Spherical computational domain. S is the poynting vector, representing the direction of energy flow. (II) Surface average of Electric field norm for a dingle gold nanohelix illuminated by (a) LCP with varying OAM (l=0,+1,-1), (b) RCP with varying OAM (l=0,+1,-1)

wavelength over a range of 600nm *i.e.* from 500nm to 1100nm.

4 RESULTS AND DISCUSSION

The results are catogerized into two parts. We started with a single gold nanohelix and proceeded further with multiple helices arranged in a particular fashion. A total of six sets of data is collected with vaying j values. We collected normalized Electric field plots to see the localized field enhancement and absorption spectra to estimate the CD value.

4.1 Single Helix

A single helix is placed in a spherical domain with a 200 nm radius and is capped with a 100 nm thick shell of PML as shown in Fig.2(I). To understand the localized field enhancement, we plotted normalized electric field by varying total angular momentum. As a control experiment, we chose an achiral object *i.e.* sphere (50nm gold nanosphere) and showed that there is no enhancement of CD. When the incident light carries OAM, the local field, generated by oscillating surface electrons, changes in response to the value of the total angular momentum. The coupling of the field to the elementary transverse surface plasma oscillations (i.e. collective oscillations of the surface electrons in the cross-sectional plane of the helix wire) is enhanced when the absolute value of j equals 2. The resonance reaches minimum value when the j equals 0 *i.e.* RCP LG_0^{-1} or LCP LG_0^{+1} (see Fig.2(II) a,b). In the same way, to monitor the absorption strength of helix, we have plotted resistive losses (Fig.3-IIa). The difference between resistive losses yield the CD spectra. The results clearly show an order of magnitude enhancement in the spectra (540 nm). It is important to note, that numerical orietational averaging [3], together with calculations, performed for an off-center helix [3], confirmed that the observed enhancement of the CD signal is neither a result of the alignment of the helix with respect to the beam axis, nor a computaional artifact.

4.2 Array of helices

For the analysis of the interaction of structured light with an array of helices, we used 12 nanohelices with center-to-center seperation of 50 nm. The results are presented in Fig. 3. The coupling between the elements of the array produces particularly strong and sharp resonance features in the spectra that can be manipulated in a switch-on/switch-off manner by changing the value of the total angular momentum quantum number. The extent of the coupling between helices is greatly affected by the value of the total angular momentum. The CD spectra for the analyzed array (Fig.3-IIb) demonstrate a great advantage of OAM-based light-matter coupling, in accord with the above analysis of an array of non-interacting helices, *i.e.* isolated helix case.

5 APPLICATIONS

Addition of OAM to the light not only enhances the resonance but also brings additional degrees of freedom that

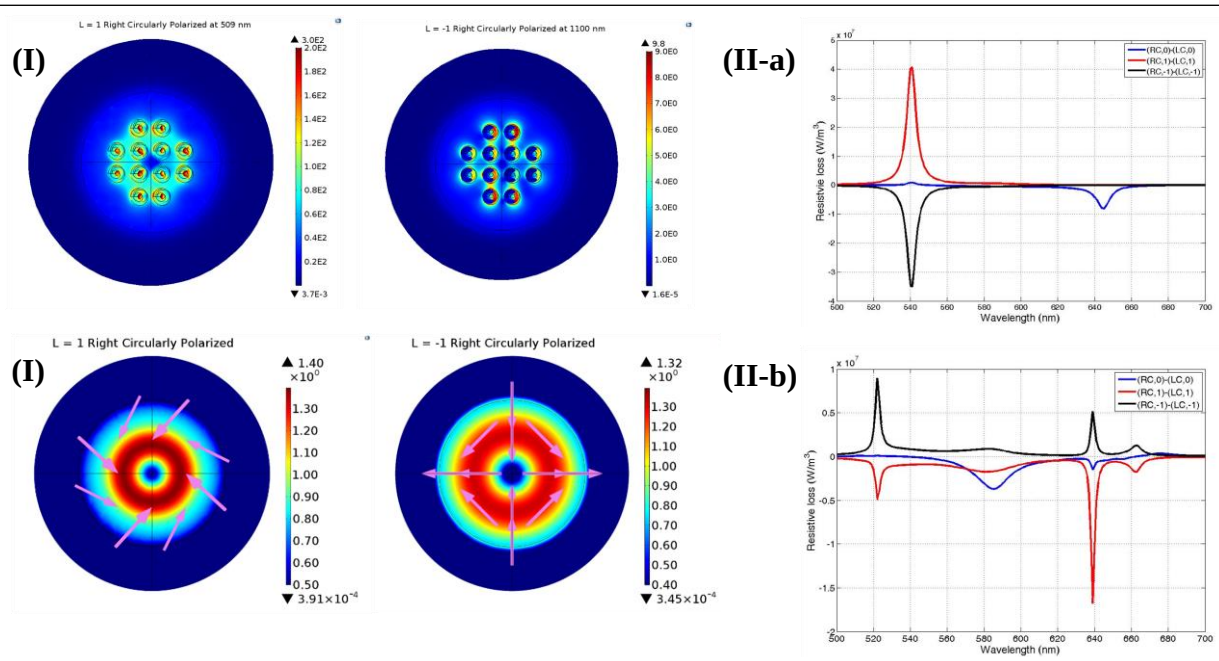

Figure 3: (I) coupling of multiple nanohelices when light with RCP and varying OAM (l = +1 and -1) is incudent on the array of multihelices (II) CD spectra obtained by taking the difference of resistive losses (absorption) (a) single gold nanohelix (b) multiple gold nanohelices seperated by 50nm.

is tunability. A key feature of this interaction is the ability to manipulate the coupling of light that has a spatially structured wavefront with these metasurfaces to achieve a manifold of sharp and significantly enhanced hybridized plasmon resonances [15]. The capability of enhancement as well as tunability can be leveraged for applications in biosensing, surface enhanced infra-red absorption, surface enhanced raman spectroscopy, laser communications, optical computing, near-zero and negative refractive index metamaterials etc [16]. Metasurfaces can revoltionize the world of science and technology with cutting-edge applications like metamaterial antenna, subwavelength photolithography, and so on. One of the key applications that can readily adapt our observations is optical computing. Basic building blocks in optical computing must operate at higher frequencies with high sensitivity and selectivity. Using metasurfaces and structured light, one can design switches and integrated electro-optic circuits. Using quasi-resonant coupling between an optically nonlinear chiral polymer and a chiral plasmonic metasurface opens up more exciting possibilities for photon spin controlled optical signal processing and beam steering. Thus, combining nanostructured chiral matter (metamaterials) with structured light (light with OAM) can bring about a synergy required for transformative advances in this area.

6 CONCLUSION

We have demonstrated that interactions between structured light and structured media facilitates unique spectral features. The synergy between them will lead to new and fundamental understanding of light-matter interactions and spur disruptive advances in myriad related technologies. While this field is still emerging, there is a proliferation of novel applications based on these complex phenomena. For the first time we have simulated the interaction of light carrying SAM and OAM with a chiral plasmonic metasurface composed of subwavelength plasmonic gold nanohelix meta-atoms. This system provides an extraordinary CD response, more than an order of magnitude greater than that obtained using conventional circularly polarized light. The selective enhancement of the resonant coupling holds promise for a gamut of applications, from enhanced bio-sensing to optical communications. Moreover, while we have considered a specific chiral metasurface configuration, the approach demonstrated here applies to a broad range of 2D and 3D plasmonic chiral media. Finally, the modeling approach that we employed can be generalized for the rational design of such media and innovative applications thereof.

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REFERENCES

1. Willner, A.E. et al. Recent advances in high-capacity free-space optical and radio-frequency communications using orbital angular momentum multiplexing. *Philos T R Soc A* **375** (2017).
2. Marrucci, L. et al. Spin-to-orbital conversion of the angular momentum of light and its classical and quantum applications. *J Optics-Uk* **13** (2011).
3. Reddy, I.V.A.K., Baev, A., Furlani, E.P., Prasad, P.N. & Haus, J.W. Interaction of Structured Light with a Chiral Plasmonic Metasurface: Giant Enhancement of Chiro-Optic Response. *Acs Photonics* **5**, 734-740 (2018).
4. Andrews, D.L. Structured light and its applications: An introduction to phase-structured beams and nanoscale optical forces. (Academic Press, 2011).
5. Beijersbergen, M.W., Coerwinkel, R.P.C., Kristensen, M. & Woerdman, J.P. Helical-wavefront laser beams produced with a spiral phaseplate. *Opt Commun* **112**, 321-327 (1994).
6. Heckenberg, N.R., Mcduff, R., Smith, C.P. & White, A.G. Generation of Optical-Phase Singularities by Computer-Generated Holograms. *Opt Lett* **17**, 221-223 (1992).
7. Yu, N.F. et al. Light Propagation with Phase Discontinuities: Generalized Laws of Reflection and Refraction. *Science* **334**, 333-337 (2011).
8. O'Dwyer, D.P. et al. Generation of continuously tunable fractional optical orbital angular momentum using internal conical diffraction. *Opt Express* **18**, 16480-16485 (2010).
9. Moreno, I., Sanchez-Lopez, M.M., Badham, K., Davis, J.A. & Cottrell, D.M. Generation of integer and fractional vector beams with q-plates encoded onto a spatial light modulator. *Opt Lett* **41**, 1305-1308 (2016).
10. Shitrit, N., Bretner, I., Gorodetski, Y., Kleiner, V. & Hasman, E. Optical spin Hall effects in plasmonic chains. *Nano Lett* **11**, 2038-2042 (2011).
11. Oh, H.S. et al. Chiral poly(fluorene-alt-benzothiadiazole) (PFBT) and nanocomposites with gold nanoparticles: plasmonically and structurally enhanced chirality. *J Am Chem Soc* **132**, 17346-17348 (2010).
12. Huang, G.S. & Mei, Y.F. Helices in micro-world: Materials, properties, and applications. *J Materiomics* **1**, 296-306 (2015).
13. Karimi, E. et al. Generating optical orbital angular momentum at visible wavelengths using a plasmonic metasurface. *Light-Sci Appl* **3** (2014).
14. Etchegoin, P.G., Le Ru, E.C. & Meyer, M. An analytic model for the optical properties of gold. *J Chem Phys* **125** (2006).
15. Prodan, E., Radloff, C., Halas, N.J. & Nordlander, P. A hybridization model for the plasmon response of complex nanostructures. *Science* **302**, 419-422 (2003).
16. Franke-Arnold, S., Allen, L. & Padgett, M. Advances in optical angular momentum. *Laser Photonics Rev* **2**, 299-313 (2008).