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Numerical investigation of far-field circular dichroism and local chiral response of pseudo-chiral meta-surface with FEM

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Abstract. Circular dichroism spectroscopy is a sensitive and widely applied technique to detect chiral molecules. Recent studies have shown high prospects for plasmonic metasurfaces of pseudo-chiral nano-resonators in enhancing chiral sensitivity. Here we study the far-field circular dichroism for gold U-shaped metasurfaces by calculating Mueller matrix elements with the Finite element method and investigate its response in light of the near field electric energy and optical chiral density.

1 Introduction

Chirality is a key structural property of molecules in biochemistry, that makes them non-superimposable onto their mirror images. Chiral molecules play a widespread role in nature, and their highly sensitive optical detection is a major challenge in life science. The pair of mirror image chiral molecules, called enantiomers, exhibit different chemical and biological properties depending on their handedness, which they unfold by interacting with other chiral objects, such as a circularly polarized light. The enantiomers show preferential absorption to left and right circular polarization, thereby revealing their presence and handedness. Circular Dichroism (CD) spectroscopy is a relevant and well-established method to quantify the electromagnetic chirality of enantiomers by measuring their differential absorption of opposite circularly polarized incident light.

Circular dichroism of a molecule (considered as an electric dipole) can be expressed as [1]

$$CD \propto A^{+}(\mathbf{r}) - A^{-}(\mathbf{r})$$

$$CD \propto \frac{\omega}{\epsilon_0}\alpha''_{ee}[U_e^{+}(\mathbf{r}) - U_e^{-}(\mathbf{r})] + \frac{2}{\epsilon_0}\alpha''_{em}[C^{+}(\mathbf{r}) - C^{-}(\mathbf{r})] \tag{1}$$

where, $A^{\pm}(\mathbf{r})$, $\mathrm{U}_{\mathrm{e}}^{\pm}(\mathbf{r})$, $\mathrm{C}^{\pm}(\mathbf{r})$ are the absorptions, electric energy densities and optical chiral densities of a randomly oriented molecule at a position $\mathbf{r}$ by two opposite circularly polarized beams denoted by the superscript $\pm$, respectively. α''_{ee}, α''_{em}, ω and ϵ_0 denote the imaginary part of the electric polarizability, imaginary part of the chiral polarizability, frequency of the incident light, and the dielectric permittivity of free space, respectively. The second part of equation (1) is attributed to the chiral absorption of the molecule, which is orders of magnitude weaker compared to the background achiral absorption, characterized by the first part of the equation, due to the intrinsic small values of α''_{em}. Therefore, detection of chiral absorption requires high sample concentration and long measurement time. The interaction of chiral molecules with electro-magnetic wave can be characterized by local chiral density C($\mathbf{r}$), expressed as

$$C^{\pm}(\mathbf{r}) = \frac{\omega}{2c^2} Im\{\boldsymbol{E}^{\pm}(\mathbf{r})^{*}\boldsymbol{H}^{\pm}(\mathbf{r})\} \tag{2}$$

Therefore, the detection sensitivity can be improved by generating strong local electro-magnetic fields. Many chiral and pseudo-chiral plasmonic nanostructures [2] have been designed to enhance the CD response, among which U-shaped scatterers have been of great importance due to their magneto-electric coupling. Recent studies have also established the relevance of dielectric achiral metasurfaces in boosting local C [1]. Previous studies have experimentally quantified the magneto-electric coupling of a single U-shaped scatterer in light of the polarizability tensors [3]. However, far-field CD as a function of the near field electromagnetic responses has not yet been investigated for a metasurface of such split-ring scatterers.

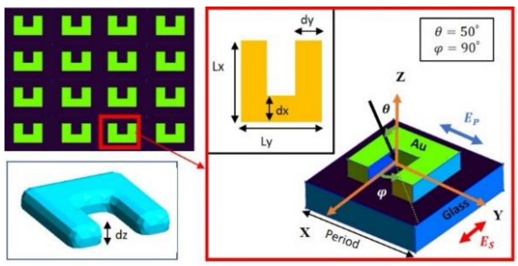

Figure. 1. Schematic of the meta-surface with U-shaped resonators with closer view of it at the inset. The parameters are $L_x = 232\text{nm}, L_y = 210\text{nm}, d_x = 90\text{nm}, d_y = 74\text{nm}, d_z = 40nm$, $Period = 390nm$. The corners of the U have been rounded off to avoid sharp resonances. The resonators are embedded in a

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glass substrate. The polar and azimuthal angles of incidence are shown with respect to the co-ordinate axes.

In this work, we numerically study the local electric energy density $U_e^{\pm}(\mathbf{r}) = \frac{\epsilon_0}{2}|E^{\pm}(\mathbf{r})|^2$ and chiral desntiy responses of gold U-shaped resonators and investigate the far field CD by computing the Mueller matrix of the metasurface.

2 Numerical analyses

The far-field optical response of a medium can be completely described by its 4x4 Mueller matrix. In this study, we calculate the Mueller matrix elements using a well-suited formulation of the Finite Element Method (FEM) [4]. This formulation rigorously addresses the vector diffraction problems by periodic crossed gratings of U-shaped and nano pillar resonators, and replaces it with an equivalent radiation problem with sources localized inside the resonator itself. Figure 1 shows the structure of the U-shaped nano-resonators. The Mueller matrix elements are calculated in the visible wavelength range (400–1200nm), at polar angle of incidence θ_0 = 0° and 50° and an azimuthal angle of incidence φ_0 = 90°, that is, along the lower arm of U. The simulations were performed for electric field components parallel (p) and perpendicular (s) to the plane of incidence containing the lower arm of the U (y-z plane). The amplitude reflection coefficients and Mueller matrix elements are calculated in the far-field by taking only the specular component of reflection into account. The local electric energy density and chiral density are calculated by averaging $\Delta U_e = {U_e}^+(\mathbf{r}) - {U_e}^-(\mathbf{r})$ and $\Delta C = C^+(\mathbf{r}) - C^-(\mathbf{r})$ over a 10nm box volume surrounding the U resonator, illuminated by right (+) and left (-) circularly polarized light.

3 Results and discussion

The normalized Mueller matrix is denoted as $[m_{ij}]$ where $m_{ij} = \frac{M_{ij}}{M_{11}}$, for $i, j = 1, 2, 3, 4$. Our study focuses on investigating the element m_{14} as it corresponds to circular dichroism. The m_{14}, ΔU_e, and ΔC spectra of the gold U metasurface for normal and oblique incidence are shown in Figures 2(a), 2(b) and 2(c), respectively. A qualitative comparison of the CD, ΔU_e and ΔC spectra indicates that for normal incidence, the symmetry properties of ${U_e}^+(\mathbf{r}) = {U_e}^-(\mathbf{r})$ and $C^+(\mathbf{r}) = -C^-(\mathbf{r})$ hold under parity operation, which, in this case, is equivalent to applying left and right circularly polarized light. Therefore, the resonators behave as achiral objects in normal incidence, without any coupling between electric and magnetic fields, leading to vanishing ΔU_e. In this case, CD can be expressed solely as a function of ΔC, which is significantly small coming only from the chirality of the incident field, thereby leading to nearly vanishing CD. On the other hand, for oblique incidence, ΔU_e becomes the main contributing factor in CD. In this case, as the magneto-electric coupling comes into play, the scattered electric field is enhanced as a function of the incidence angle [3].

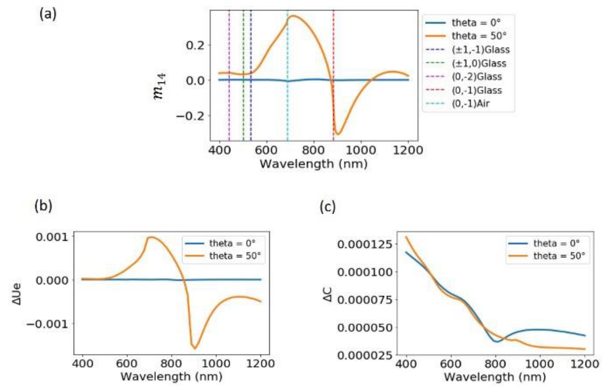

Figure. 2. FEM simulations of (a) far field CD (m_{14}) for the gold U metasurface, (b) near field electric energy density and (c) optical chiral density spectra averaged over a box volume around a unit U resonator for normal (blue) and oblique (yellow) incidence. The vertical lines in (a) represent Rayleigh lines corresponding to the sharp edges of the spectra.

This enhancement of field with angular dependence appears in square in ΔU_e spectra, inducing an extrinsic chirality. However, in presence of a chiral medium, the desired intrinsic chiral signal from the molecules will be dominated by this large extrinsic chiral absorption from the U resonators due to the much higher values of ΔU_e compared to ΔC. The extrinsic chirality would also produce CD signals for achiral molecules with $\alpha''_{em} = 0$ [1]. Therefore, it would be interesting to design the metasurface in a way to remove the contribution from ΔU_e, and still benefit from the enhanced scattered field due to magneto-electric coupling in oblique incidence.

In the next steps, we will investigate new metasurface designs with U resonators in order to describe CD solely as a function of the intrinsic chiral density created by the resonators. The local chiral density will then be optimized by tailoring the geometrical parameters of the resonators. We will also experimentally investigate the polarimetric properties and sensitivity of the designed metasurface to an ambient medium to compare with other relevant metasurfaces.

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