Spatially Resolved Enhancement of Chirality in Planar and 3D Chiral Metamaterials

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Abstract

We visualize the enhancement of optical chirality in the near-field regime of chiral metallic metamaterials. Different planar and three-dimensional geometries are analyzed. We show that two-dimensional structures are more suitable for practical applications as it is easier to access the superchiral fields, while three-dimensional structures show higher enhancement of optical chirality.

1. Introduction

In recent years, a new class of metamaterials has gained a lot of interest: chiral metamaterials, which show a polarization-dependent interaction with circularly polarized light (CPL) due to their inherent structural handedness. Circular dichroism and optical activity, which are several orders of magnitude higher compared to typical values of chiral molecules or liquid crystals, have been reported [1].

Up to now, most research has been dedicated to the far-field response; the near-field behavior is basically unknown. We close this gap by investigating the nearby electromagnetic field of planar as well as three-dimensional metallic metamaterials and show that—similar to the field-enhancement effect—a chiral nanostructure enhances the so-called *local optical chirality*

$$C(\mathbf{r}) = \frac{\varepsilon_0}{2}\mathbf{E}(\mathbf{r}) \cdot \nabla \times \mathbf{E}(\mathbf{r}) + \frac{1}{2\mu_0}\mathbf{B}(\mathbf{r}) \cdot \nabla \times \mathbf{B}(\mathbf{r})\,, \tag{1}$$

which is a measure for the chirality of the electromagnetic field. It is directly correlated with the rate of excitation of a chiral molecule within the chiral field [2]. Therefore, by using fields with enhanced optical chirality (so-called *superchiral fields*), the sensibility of enantiomer sensors using circular dichroism to determine the handedness can be improved. Although this principle has been demonstrated recently [3], systematic studies of different structures are necessary to optimize such sensors.

2. Visualization of Optical Chirality Enhancement

Up to now, in circular dichroism experiments mostly the optical chirality difference between left- and right-handed CPL (C_+ and C_-) was considered. By the use of a chiral metamaterial one obtains modified values for the optical chirality $C_\pm^{\text{struct}}$ when the structure is illuminated with left- (+) or right-handed (-) CPL. To compare different structures, we calculate the normalized chirality enhancement $\hat{C}_\pm = \frac{C_\pm^{\text{struct}}}{|C_\pm|}$. In general, these values can differ in both their signs and their magnitudes. When comparing the transmittance obtained for the two incident polarizations, the difference in the response is determined by the

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change of the optical chirality enhancement

$$\Delta\hat{C} = \frac{1}{2}\left(\hat{C}_{+} - \hat{C}_{-}\right) . \tag{2}$$

This is the fundamental parameter we base our comparison on. As the enhancement is a near-field effect, the shape of the superchiral field strongly depends on the geometrical structure of the analyzed metamaterial. To obtain a general overview, we show three-dimensional maps of $\Delta\hat{C}$. From these maps one can identify regions with locally enhanced chirality change. A quantitative view can be gained by slices through the structure, which allow a color-coded visualization of $\Delta\hat{C}$.

3. Planar Metamaterials

Planar chiral metamaterials such as a metallic spiral represent a convenient way to generate superchiral fields. Compared to metamaterials with three-dimensional chirality, planar chiral metamaterials are easier to fabricate and more suitable for sensing applications.

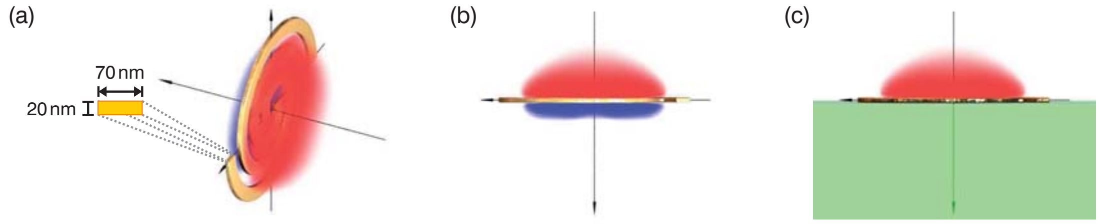

Fig. 1: Distribution of enhanced chirality change $\Delta\hat{C}$ caused by a planar gold spiral. (a) Perspective view. (b) Side view showing the clear separation between regions where the handedness of the incident light is preserved (red) or changed (blue). (c) Only one of these regions can be accessed in presence of a substrate.

The enhanced chirality change $\Delta\hat{C}$ for a two-armed spiral in air is shown in Figs. 1a (perspective view) and 1b (side view). One can clearly distinguish between the region at the front, where a positive enhancement (red) of more then a factor of 10 is present, and the negative values (blue) at the back. They describe locations, where the optical chirality change is not only enhanced, but also the handedness of the incident light is changed.

For sensing applications which make use of the superchiral fields it is important to access only one of the polarities. Otherwise, the overall response would be decreased. This access scheme is naturally supported by planar structures, as they are commonly fabricated on top of a substrate which blocks the access to one of those regions (cf. Fig. 1c).

4. Three-dimensional Structures

As a first three-dimensional structure, we study the so-called stereometamaterial, which consists of two split-ring resonators (SRRs) with the twist angle Θ as a free parameter (cf. Fig. 2a) [4]. The chiroptical response (illustrated by the maximum and minimum values appearing for the circular transmittance difference) shows a complex dependence on Θ due to the near-field coupling of the SRRs, which is demonstrated in Fig. 2b: besides the trivial szenarios at $\Theta = 0\,^\circ$ and $\Theta = 180\,^\circ$, one additional setting with minimum response occurs at $\Theta = 55\,^\circ$, which coincides with the minimum spectral distance between the hybridized plasmon modes predicted in [4]. Figure 2c shows the distribution of the enhanced chirality change. Obviously, the regions with different signs cannot be isolated as easily as for the two-dimensional example shown in Sect. 2. Note that we used the exact same geometry parameters as well as the embedding material as in Ref. [4], but a Drude model for gold. Hence, the minimum angle of $55\,^\circ$ is specific for the given size and distance ratios.

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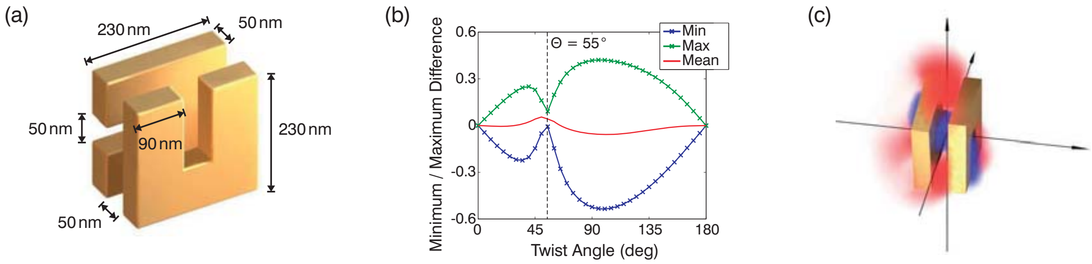

Fig. 2: Chiroptical response of the stereometamaterial. (a) Dimensions of the structure. (b) Extreme values of the transmittance difference between left- and right-handed circularly polarized light for different twist angles Θ. For $\Theta = 55°$ one finds a non-trivial szenario with minimum chiroptical response. (c) Enhanced chirality change for $\Theta = 110°$ (angle with maximum chiroptical response).

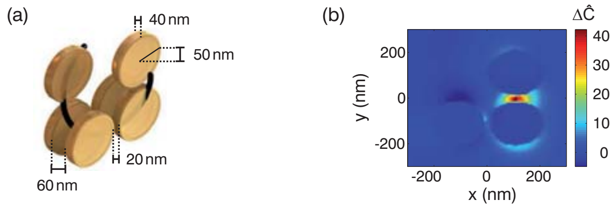

Fig. 3: (a) Plasmonic gold oligomer with helix-like geometrical chirality. (b) Slice through the chirality change map perpendicular to the propagation direction of the incident light at $z = -38$ nm. The metamaterial shows an enhancement of more than a factor of 40.

Additionally, we analyze chiral oligomer structures which mimic a helix (cf. Fig. 3a). Compared to real helices, these oligomers can be fabricated by well-established processes such as electron-beam lithography. We show an optimized structure which enhances $\Delta\hat{C}$ by a factor of more than 40.

5. Conclusion

In conclusion we have visualized the change of the optical chirality enhancement for different chiral metamaterials. The highest values were obtained for three-dimensional plasmonic oligomer structures. Two-dimensional metamaterials have an advantage in practical applications, as they are easier to fabricate. Additionally, the access to regions with unchanging polarity of the enhancement is convenient.

The demonstrated analysis is of fundamental importance when designing chiral structures for future applications like ultra-sensitive enantiomer sensors. For an experimental verification of the calculated results, one could use the interaction of chiral fields with chiral molecules. However, to obtain quantitative results with high spatial resolution, novel experimental techniques have to be developed.

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