



## *Optical Chirality Enhancement in Twisted Arrays of Plasmonic Nano-rods*

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### **ABSTRACT**

An important property of electromagnetic fields, which arises from the interaction between fields and chiral molecules, is called optical chirality. By enhancing this field property, while maintaining constant input power, we are able to increase absorption of circularly polarized light by chiral molecules of a certain handedness. This enhancement is achieved through the use of achiral plasmonic nano-particles in conjunction with the twisted metamaterials. Optical chirality enhancement (OCE) has an important application in sensing enantiomers of chiral molecules. Here, we present a preliminary scheme to measure enantiomeric excess in mixtures of chiral molecules using OCE boosted by twisted metamaterials. This scheme does not require measurement of a frequency shift in the circular dichroism response.

### **KEYWORDS**

Enantiomeric Excess, Optical Chirality, Plasmonics, Twisted Metamaterial.

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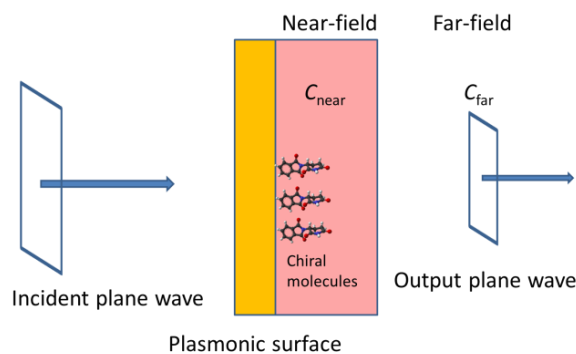
## 1- INTRODUCTION

Since Louis Pasteur's discovery [1] that tartaric acid can have two forms with opposite senses of rotation; the interaction between electromagnetic waves and chiral molecules has interested many scientists and engineers. Chiral objects lack the mirror symmetry, in other words, one cannot map the object to its mirror image by simply translating or rotating the object. The object and its mirror image form an enantiomeric pair. In nature, chiral molecules which have an active role in biology are often homochiral, which means that one enantiomer outweighs the other one. However, in manufacturing artificial bioactive molecules, because of the symmetries in the manufacturing process, often the final product is a mixture of both enantiomers. Although Enantiomers, made of the exact same constituents, can have drastically different chemical properties.

Distinguishing enantiomers is important because a pair of such molecules have exactly the same chemical constituents but with the opposite handedness. These chiral isomers can have significantly different effects and toxicity when entered the human body. This makes the ability to separate enantiomer pairs extremely important for the pharmaceutical industries, since one handedness of a molecule can be used to cure a certain illness, and the other handedness can have serious side effects. The most infamous of these molecules is the drug thalidomide. While one enantiomer cures morning sickness in pregnant women and has other beneficial effects, the other enantiomer causes malformation of limbs in the babies born [2].

Therefore, manufacturing single-enantiomer drugs is an important part of the pharmaceutical industry which amounts to billions of dollars of market value per year [3]. In order to reduce costs in the manufacturing process, it is essential to have cheap, accurate and sensitive measurement methods to discriminate between enantiomers of a single molecule. A class of methods has been developed to distinguish between chemical enantiomers through their optical activity. As an example, circular dichroism spectroscopy is heavily used to detect the chiral biomolecules [4]. The interaction between chiral molecules and electromagnetic fields, which results in circular dichroism, is entirely dependent on the optical chirality of the field.

Chirality of electromagnetic fields (optical chirality) was first studied as a conserved quantity in free-space propagation [5]. This concept was revisited recently because of its importance in the interaction between chiral molecules and electromagnetic fields [6]. Optical chirality is also important from a symmetry point of view [7], where it fills the gap for a pseudo-scalar (a scalar with odd symmetry under mirror symmetry) quantity with even time reversal symmetry. Since the optical chirality affects the amount of absorption of electromagnetic energy by a single chiral molecule, it shows promising potentials for manipulating chiral molecules and also differentiating between enantiomers of chiral molecules [8], [9].



**Fig. 1. The setup for enhancing optical chirality. A circularly polarized plane wave illuminates the metamaterial structure with plasmonic inclusions. On the right-hand side of the structure, there is a region dominated by near-field phenomena where optical chirality is enhanced compared to the optical chirality at the far-field region. The enantiomers of chiral molecules should be placed in the near-field zone.**

By drawing analogy from near field enhancement in plasmonic materials, one can conceive the idea of enhancing the optical chirality on the surface of plasmonic materials [10]. This enhancement in the optical chirality can be used to greatly enhance the interaction between chiral parts of the electromagnetic field and chiral molecules/materials [11]. Therefore, it can be used to increase our ability to measure/manipulate chiral structures using electromagnetic radiation. To gain nonzero net optical chirality, we need to break the mirror symmetry of the structure. This can be done by introducing twist in the structure. Although these twisted structures have been studied extensively for their enhanced optical activity [12] and their modal properties [13], here, we investigate their near-field chirality enhancement properties.

This paper is an extension to the work presented in [14]. In the next section, we briefly review the concept of optical chirality. A discussion of the interaction between chiral molecules and electromagnetic fields is presented in the section III. Section IV is devoted to the discussion of the enhancement in the optical chirality, especially in the near-field of plasmonic nano-particles. A new measurement scheme is proposed and discussed in section V and finally section VI provides some concluding remarks.

## 2- OPTICAL CHIRALITY

Lipkin introduced a time-even pseudo-scalar quantity called optical chirality as [5]

$$C = \frac{\epsilon_0}{2} \mathbf{E} \cdot \nabla \times \mathbf{E} + \frac{1}{2\mu_0} \mathbf{B} \cdot \nabla \times \mathbf{B} \quad (1)$$

where  $\epsilon_0$  and  $\mu_0$  are the free-space permittivity and permeability, respectively.  $\mathbf{E}$  and  $\mathbf{B}$  are the time dependent electric and magnetic field vectors and  $C$  is the optical chirality as a function of time and space variables. This time-dependent quantity can be expressed in the time-harmonic case, as

$$\tilde{C} = -\frac{\epsilon_0 \omega}{2} \text{Im}(\tilde{\mathbf{E}} \cdot \tilde{\mathbf{B}}^*), \quad (2)$$

where  $e^{i\omega t}$  time dependence is assumed and suppressed in the formulation. The above equation is derived in a source-free region. The tilde above field quantities represents the time-harmonic nature of these vectors.

Originally, no physical meaning was attached to the above quantity. However, it is shown that it satisfies a conservation law in the following form [6].

$$\frac{\partial C}{\partial t} + \frac{1}{\mu_0} \nabla \cdot \Psi = -\frac{1}{2} (\mathbf{j} \cdot \nabla \times \mathbf{E} + \mathbf{E} \cdot \nabla \times \mathbf{j}) \quad (3)$$

where  $\mathbf{j}$  is the current density as a function of time, and the flux of chirality  $\mathbf{F}$  is defined as

$$\Psi = \frac{1}{2} \mathbf{E} \times (\nabla \times \mathbf{B}) - \frac{1}{2} \mathbf{B} \times (\nabla \times \mathbf{E}), \quad (4)$$

and  $\mathbf{j}$  is the current density as a function of time. Equation (3) is similar to other conservation laws such as Poynting theorem. If one compares equation (3) and the Poynting theorem, one finds that  $C$  acts analogously to the energy density,  $\Psi$  to the Poynting vector and the right-hand side of equation (3) to the source term. This justifies the claim that optical chirality  $C$  can be thought of as the density and  $\Psi$  as the flux of a second order quantity.

The time averaged chirality flux in the time-harmonic case has the following form,

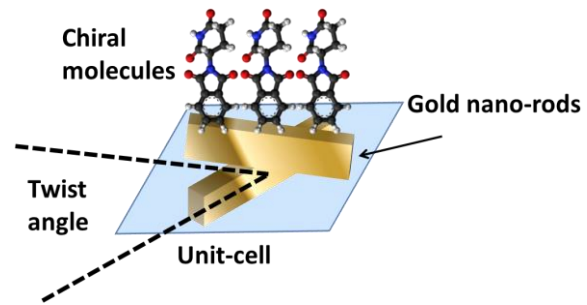
$$\langle \tilde{\Psi} \rangle = \frac{1}{4} \text{Re} \left[ \tilde{\mathbf{E}} \times (\nabla \times \tilde{\mathbf{B}}^*) - \tilde{\mathbf{B}} \times (\nabla \times \tilde{\mathbf{E}}^*) \right]. \quad (5)$$

The above equation can be simplified with the free space Maxwell's equation. In this way, one can rewrite (5) as,

$$\langle \tilde{\Psi} \rangle = \frac{\omega \mu_0}{4} \text{Im} \left[ \epsilon_0 \tilde{\mathbf{E}} \times \tilde{\mathbf{E}}^* + \mu_0 \tilde{\mathbf{H}} \times \tilde{\mathbf{H}}^* \right]. \quad (5)$$

The above equation has been shown to be closely related to the spin and orbital angular momentums of light. This flux term is partly responsible for the force exerted on a chiral particle [9] by chiral electromagnetic fields.

The symmetry properties of the abovementioned quantities are also of interest. While the energy density is even under parity symmetry, optical chirality  $C$  has odd parity symmetry. In other words,  $C$  changes sign under a point reflection. In this sense, optical chirality behaves according to the expectations for a quantity corresponding to chirality. From equation (4), it is clear that  $\Psi$  has an even parity symmetry, and the right-hand side of equation (3) has an odd parity symmetry. Another important symmetry operation is the time reversal symmetry. While  $C$  has an even time reversal symmetry, both  $\Psi$  and the source terms have odd time reversal symmetries. From these symmetry considerations, it is obvious that in order to generate electromagnetic waves with non-zero optical



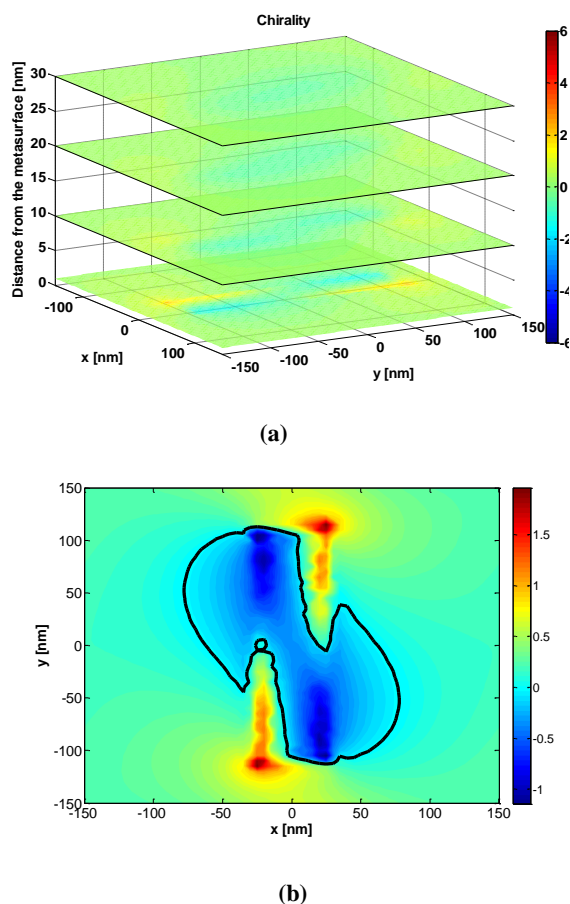
**Fig. 2.** Two gold nano-rods are placed on top of each other. The upper one is rotated using a twist angle. The whole structure is repeated in a two dimensional array. The chiral molecules are placed on the top layer, where the optical chirality is enhanced and therefore the interaction between chiral parts of the field and molecule is amplified.

chirality, we need to break the mirror symmetry of the structures in the spatial domain.

As a simple example of optical chirality, one can show that for a linearly polarized plane waves the optical chirality is always zero and for right-handed (RCP) and left-handed (LCP) circularly polarized plane waves the chirality is  $\pm \omega \epsilon_0 |E_0|^2 / c_0$ , where  $E_0$  is the electric field amplitude, and different signs are for opposite handedness (positive for the LCP waves). Although circularly polarized waves exhibit the maximum possible optical chirality for plane waves, it is nevertheless possible to obtain higher values for the optical chirality with a combination of propagating and evanescent waves. This is due to the fact that  $C$  is a quadratic quantity in the fields and therefore, the optical chirality of two plane wave simultaneously propagating through some part of space is not simply the sum over optical chirality of each plane wave separately. In order to make this point more clear, consider two plane waves, one propagating in the  $+z$  direction with its electric field ( $E_0$ ) linearly polarized in the  $x$  direction and its magnetic field ( $B_0$ ) in the  $y$  direction. Obviously, the optical chirality of this plane wave is zero. Now, if we introduce another plane wave propagating in the  $+z$  direction with its electric field ( $E_0$ ) in the  $y$  and its magnetic field ( $B_0$ ) in the  $-x$  directions, the optical chirality of this second wave is also zero. However, the superposition of these two linearly polarized plane waves will produce a circularly polarized wave with a nonzero optical chirality. In the following sections, we will be searching for a way to obtain fields with an optical chirality that surpasses the optical chirality of circularly polarized plane waves.

### 3- INTERACTION OF CHIRAL FIELDS WITH CHIRAL MEDIUM

The amount of loss in a chiral material is related to the product of the imaginary part of material chirality ( $\text{Im} \kappa$ ) and the optical chirality (i.e. power loss due to chirality is proportional to  $\text{Im}[\kappa] \text{Im}[\tilde{\mathbf{E}} \cdot \tilde{\mathbf{B}}^*]$ ) [15], where  $\kappa$  is the chirality in Tellegen's notation. Therefore, for a chiral material, if one could increase the optical chirality while



**Fig. 3.** The optical chirality of the fields for the structure of Fig. 2 when there are no chiral molecules and the twist angle is zero. The incident field is a RCP plane wave illumination the structure from below. Average enhancement factor for  $z=1\text{nm}$  is around 3. (a) Chirality on different distances from the structure and (b) chirality at  $z=5\text{nm}$ . The solid line depicts the boundary between positive and negative optical chirality. For LCP plane wave the results are mirror image of above results with inverted sign, as expected from mirror symmetry argument.

keeping the incident power constant, it would be possible to increase the interaction between the chiral matter and chiral electromagnetic field at a constant power level.

Another way to demonstrate this effect is by considering the amount of energy absorbed by a single particle. If we model the particle using its electric and magnetic dipole moments and define the magneto-electric dipole polarizability ( $\alpha_{em}$ ) as the term connecting electric dipole moment to the magnetic field and the magnetic dipole moment to the electric field (i.e.  $\mathbf{p} = \alpha_{ee}\mathbf{E} + \alpha_{em}\mathbf{B}$  and  $\mathbf{m} = -\alpha_{em}\mathbf{E} + \alpha_{mm}\mathbf{B}$  where  $\mathbf{p}$  and  $\mathbf{m}$  are electric and magnetic dipole moments), it is possible to show that the amount of absorbed energy due to chirality is proportional to  $\text{Im}\alpha_{em}C$  [6].

By enhancing the optical chirality in the vicinity of chiral molecules, the interaction between electromagnetic fields and the molecules is improved. This will result in

higher absorption of electromagnetic energy by molecules with an appropriate chirality and lower absorption of electromagnetic energy by the molecules with an opposite handedness. In addition, it is anticipated that because of this enhanced interactions, the optical forces exerted on chiral molecules become stronger in the right circumstances. One can use this enhanced optical forces to manipulate chiral molecules more effectively. However, the forces exerted on chiral particles by optical fields are not addressed here and will be discussed in detail in a future work.

Optical chirality enhancement (OCE) is obtained by placing materials with a suitable shape and structure in the transmission path of the electromagnetic wave. Interaction between these structures and the wave will usually result in a complicated electromagnetic field near the structure. By properly designing the structure and deploying appropriate materials, one can obtain larger optical chirality in the proximity of the structure. There are different ranges of materials that can be used to obtain optical chirality enhancement. However, since plasmonic metals have been shown to achieve ultra-high near-field enhancement [16], it is reasonable to expect high OCE near plasmonic particles as well.

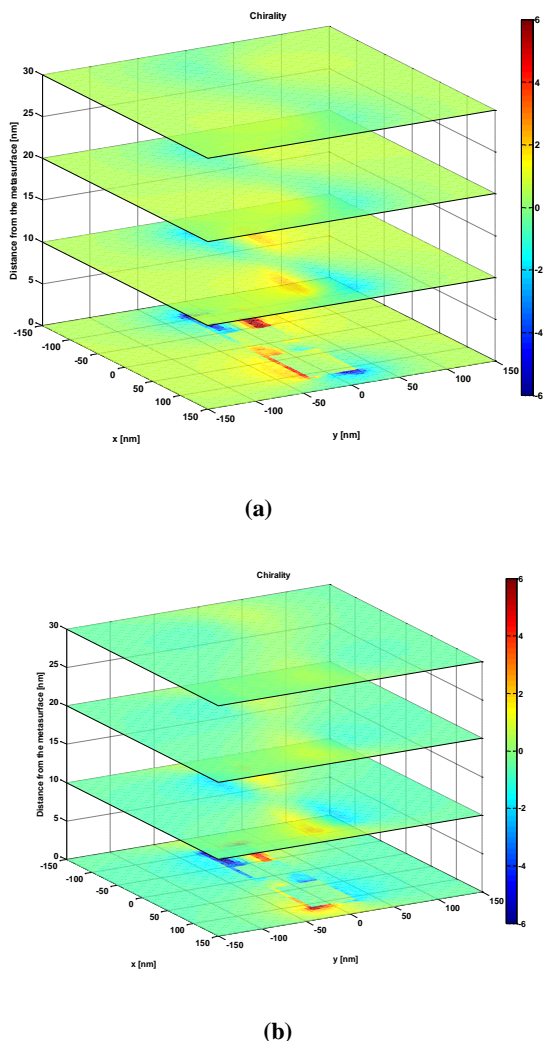
In order to study the amount of OCE near the surface of a plasmonic particle, we define the OCE factor as the ratio between the optical chirality in the far-field of a plasmonic surface and the optical chirality in the near vicinity of the surface. Explicitly, we use the following formula for the chirality enhancement.

$$K = C_{near} / C_{far} \quad (6)$$

when the structure is illuminated using a plane wave. Far-field chirality is calculated where the fields can be approximated well enough using a single plane wave at the output side of the plasmonic surface, and  $C_{near}$  is calculated in the near-field zone of the enhancing surface. The setup for this calculation is shown in Fig 1.

The OCE factor as defined in (6) captures the amount of increases in the absorption of the wave energy due to the interaction between the chiral parts of the field and molecules, compared to when the molecules are not located in a chirality enhanced region. Obviously, this enhancement factor depends on the location of the molecules, and we expect that as we increase the distance between the molecules and the surface, the amount of the enhancing factor decreases. The OCE factor usually depends not only on the distance between the plasmonic particle and the chiral molecule, but also on the relative positioning between the chiral molecule and the plasmonic particle. Since it is usually difficult to control the relative placement of the molecules on the plasmonic particles, it is desirable to achieve high optical chirality irrespective of the positioning of the molecule on the plasmonic particle.





**Fig. 4.** The optical chirality of the fields for the structure of Fig. 2 when there are no chiral molecules and the twist angle is 90 degrees. The incident field is a (a) RCP and (b) LCP plane wave illumination the structure from below. Optical chirality is much stronger in this case compared to the case shown in the Fig. 3.

#### 4- ENHANCEMENT FACTOR IN TWISTED METAMATERIALS

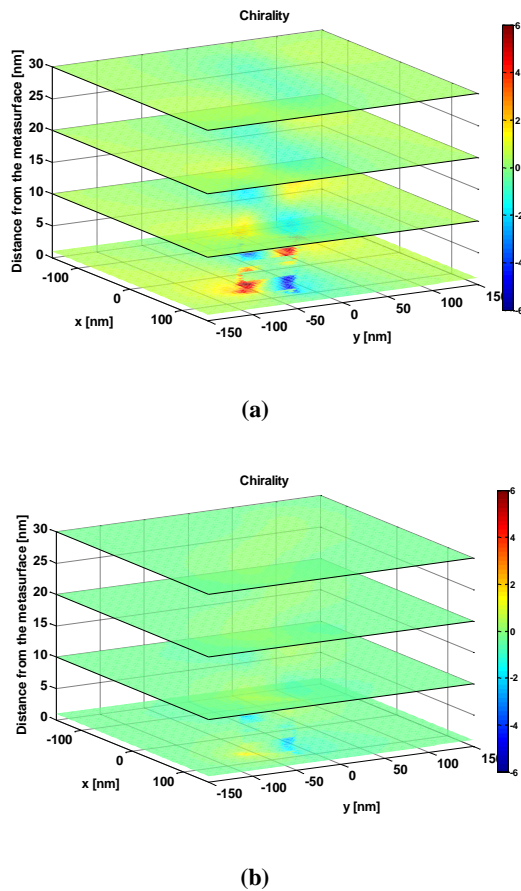
Enhancement in the optical chirality can be achieved in different ways. One can use an intrinsically chiral plasmonic particle (e.g. a gammadion) [17] or impose extrinsic chirality on an array of intrinsically achiral particles [18] using properties of the global structure. Here, we focus on the second method and will use achiral gold nano-rods in a twisted array to enhance the local field chirality. The structure of the resulted twisted metamaterial with chiral molecules placed on top of it is shown in Fig. 2. Since we understand the nature of propagating modes in a twisted metamaterial [13], we can analytically calculate the optical chirality of the modes on the output side of the twisted metamaterial slab. However, since in these calculations we do not take into account the near-field phenomena associated with plasmonic

materials, it is impossible to obtain an accurate description of the enhancement using the analytical model. Therefore, we will study the optical chirality and its enhancement by numerical simulations of the structure.

Using commercially available full-wave simulation software (e.g. Ansys HFSS) and previously reported optical properties of gold at terahertz frequencies [19], we can calculate the optical chirality for different structures. It is worth mentioning that in all the simulations carried out here, the material loss for gold is taken into account. At 300THz, while its refractive index is 0.229, the extinction coefficient of gold has a value equal to 6.79 [19]. This significant amount of loss, which is present in all plasmonic materials, reduces the OCE factor of the structure. Therefore, selecting the right plasmonic material for the desired frequency is essential.

As a first example, we have simulated a two-layer array of gold nano-rods with the dimensions of 50nmx40nmx220nm at 300THz. The nano-rods are placed in a square lattice. Both lattice constants are 300nm. The vertical distance between the nano-rods is 100nm center to center. The OCE factor for this structure at different distances from the top surface is shown in Fig. 3(a) for a zero twist angle. The incident wave is a right-handed circularly polarized plane wave propagating in a normal direction to the surface containing the rods. The top side of the rod is at  $z = 0$  and the first plane in the figure is at  $z = 1$ nm. Obviously, this case possesses two symmetry planes at  $x = 0$  and  $y = 0$ . Although the excitation in this case breaks the mirror symmetry (since it is circularly polarized and its mirror symmetry has an opposite handedness), these symmetry planes are responsible for the approximate mirror symmetry that is observed in the optical chirality of the structure. In addition, it is clear from the figure that at larger distances from the nano-rods, the enhancement in the optical chirality gradually dies out. Therefore, one can conclude that the OCE is essentially a near-field phenomenon arising from plasmonic resonances of the nano particles. It is important to note that although the incident wave has a pure circular polarization, through interaction with the nano-rods, both positive and negative optical chiralities are produced in the output side of the structure.

Part (b) of Fig. 3 shows the field chirality at a 5nm away from the top surface and highlights the boundary between positive and negative field chirality regions using a black line. If there were no rods present, the field chirality will have a negative sign throughout the space. However, because of the presence of the plasmonic nano-rods, at some places, the field chirality changes sign and becomes positive. If we excite the same structure with a plane wave of opposite handedness, all the results would remain the same, but with an opposite sign. Therefore, this mirror symmetric structure enhances right and left-handed field chiralities in an equal amount. In addition, it is clear from the figure that near the corners of the nano-rods, where we have sharp edges, the optical chirality reaches much higher levels compared to the more smooth parts of the center of the rod.



**Fig. 5.** The optical chirality of the structure with a twist angle of 60 degrees. Other parameters are similar to Fig. 3. The incident fields is (a) RCP and (b) LCP plane wave.

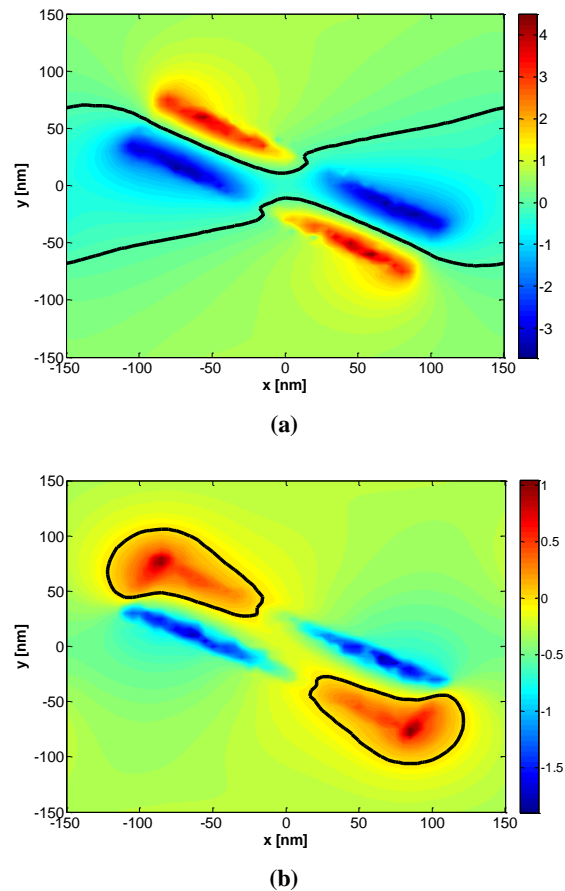
Similar calculations are carried out for different twist angles. From these calculations, one can conclude that there are always regions of enhanced field chirality near the nano-rod with different signs for the enhancement factor. The optical chirality in the case of 90 degrees rotation between the nano-rods is displayed in Fig. 3. Parts (a) and (b) of the figure are calculated using right- and left-handed circularly polarized incident plane waves. In this case, the optical chirality achieves much higher values compared to the previous case. However, as can be seen from the figure, again, we see a symmetrical response to right- and left-handed polarizations. This is expected because with 90 degrees rotations, we still have the mirror symmetries in the structure.

An approximately maximal enhancement occurs at the twist angle of 60 degrees as shown in Fig. 4 with an average OCE around 5. Part (a) and (b) show chirality for excitations with the opposite handedness. The dark black lines depict the boundary between positive and negative optical chiralities in this plane. Note that in parts (a) and (b), the colors correspond to different values of optical chirality. In this structure, RCP incident waves produce larger OCE factors for both positive and negative values of chirality. However, by examining a more detailed representation of chirality at  $z = 5\text{nm}$  in Fig. 5, it becomes

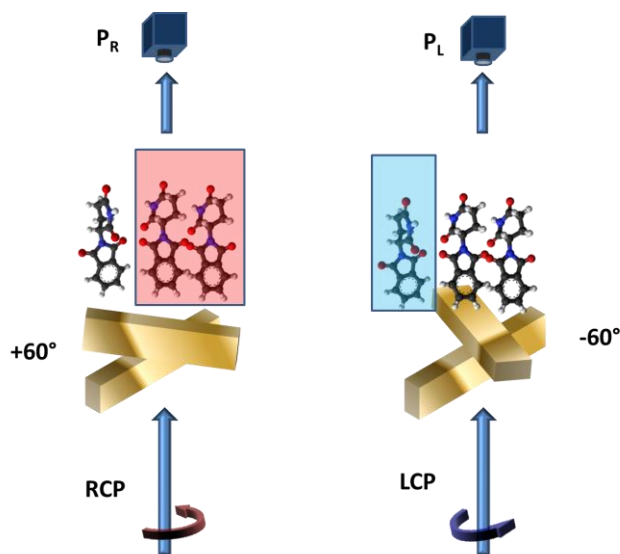
clear that the OCE is more pronounced for positive chirality in this case. In other words, positive near-field chirality has greater values, and it occupies larger surface area of the structure for RCP incident waves. For the other handedness, while negative field chirality is more prevalent, the absolute value of enhancement is not as large. This asymmetry between the two circularly polarized waves is a direct result of breaking the symmetry of the structure. It is essential for discrimination between chiral molecules with different chiralities to have the abovementioned asymmetry in the response of the structure.

### 5- ENANTIOMERIC EXCESS MEASUREMENT SCHEME

A chiral molecular sensing scheme using a similar twisted structure is proposed in [11]. The proposed scheme is based on the measurement of the frequency response of the chiral molecules placed in the vicinity of a twisted array of plasmonic nano-rods. By measuring the circular dichroism of the structure when there is no chiral molecule on top of the structure, we obtain a reference for the next measurement step. After placing the chiral molecules on top of the array, we measure the circular



**Fig. 6.** The optical chirality on a plane 5nm above the top surface of the structure of Fig. 4 with (a) RCP and (b) LCP incident plane wave.



**Fig. 7. A scheme for measuring enantiomeric excess using OCE in a twisted metamaterial with gold nano-rod inclusions. In the first measurements the electromagnetic energy is absorbed mainly by right-handed molecules and in the second case by left-handed molecules. From difference in the measured power in the two cases ( $P_R$  and  $P_L$ ), it is possible, in principle, to obtain the ratio of left and right handed molecules.**

dichroism again. It can be shown that in the second case, the position of the maximum of the circular dichroism shifts in frequency. This shift is proportional to the imaginary part of  $\kappa$ . Although this is an effective technique and can be very sensitive to small concentrations of the chiral molecules, the shift in the circular dichroism is also small, and in some cases, it might be difficult to measure this shift due to the presence of noise and other unpredictable variations in the measurement setup. In order to avoid the measurement of a shift in the frequency response, here, we propose a new scheme to measure the enantiomeric excess of a solution of chiral molecules.

From mirror symmetry considerations, it is clear that for a twist angle of  $-60^\circ$ , the OCE has exactly the same structure as shown in Fig. 5 and Fig. 6. The only difference would be the inversion of the sign of optical chirality. Based on these properties of OCE, it is possible to propose the following scheme for the measurement of enantiomeric excess (i.e. absolute difference between mole fractions of each enantiomer) based on OCE in structures with plasmonic inclusions. Consider a mixture of chiral isomers placed on top of two twisted metamaterials with opposite senses of rotation as shown in Fig. 7. The  $+60^\circ$  metamaterial has higher OCE factor when illuminated by an RCP plane wave and the  $-60^\circ$  metamaterial has a higher enhancement factor for LCP incidence.

If we illuminate these two setups with the appropriate circularly polarized plane waves, in the first setup the electromagnetic field interacts more with one group of

isomers while in the second setup it interacts with the complementary group. Therefore, the electromagnetic energy absorbed in each case depends on the relative portion of each enantiomer in the mixture. For example, a racemic mixture with equal amounts of right- and left-handed molecules absorbs both RCP and LCP equally and thus  $P_R = P_L$ , where  $P_R$  and  $P_L$  are power received by the sensors in each setup. If the mixture is made purely of right-handed molecules, there would be no absorption for the left-handed wave and  $P_L = P_0$  where  $P_0$  is the power received by the sensor when no molecule is present. On the other extreme when there are only left-handed molecules, we have  $P_R = P_0$ . For other ratios between enantiomers, it is possible, in principle, to establish a relationship between the power absorbed by the molecules in each case and the enantiomeric excess. We expect this scheme to provide a more sensitive way of measuring the abovementioned quantity, because of the chirality enhancement and stronger interaction between electromagnetic fields and molecules.

The advantage of the proposed measurement scheme is that there is no need to measure the spectrum of circular dichroism to obtain the frequency response and then find the frequency shift. It is possible to perform the measurements in a single predefined frequency and compare the observed powers in that frequency. In addition, in this measurement scheme, it is possible to find the enantiomeric excess, while in the work reported in [11], it is not clear that how the shift in the frequency response is related to the enantiomeric excess.

We have used twisted arrays of plasmonic nano-rods to predict the behavior of this measurement setup. However, it seems that if we can obtain the required OCE through other means, we can still use the same principles to perform the abovementioned measurements. For example, other non-symmetrical nano-particles such as gammadions could serve as an optical chirality enhancement platform. However, we need to modify these nano-particles to ensure that the response for right- and left-handed incident waves is not exactly similar to each other.

## 6- CONCLUSION

The concept of optical chirality enhancement is briefly discussed, and some of its implications are reported. We have numerically investigated optical chirality and its enhancement in twisted metamaterials with intrinsically achiral inclusions. The twisted metamaterial used in this paper is composed of plasmonic nano-rods made from gold. We have only used two layers of gold nano-rods in the proposed measurement scheme. Based on the properties and responses of this twisted metamaterial, a new scheme is proposed to measure the enantiomeric excess in solutions of chiral molecules. The scheme does not require a measurement of the frequency shift in the circular dichroism response of the chiral molecules. Rather it only requires measurement of optical power at a predefined frequency where the response of the twisted metamaterial is optimized.

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