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RESEARCH ARTICLE

REVISED Vortex Circular Dichroism: An experimental technique to assess the scalar/vectorial regime of diffraction

[version 2; peer review: 2 approved]

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Abstract

Background

In classical electrodynamics, light-matter interactions are modelled using Maxwell equations. The solution of Maxwell equations, which is typically given by means of the electric and magnetic field, is vectorial in nature. Yet it is well known that light-matter interactions can be approximately described in a scalar (polarization independent) way for many optical applications. While the accuracy of the scalar approximation can be theoretically computed, to the best of our knowledge, it has never been determined experimentally. Here, we introduce Vortex Circular Dichroism (VCD), an optical measurement that has the required features to assess the vectoriality of diffraction.

Methods

VCD is measured as the differential transmission (or absorption) of left and right circularly polarized vortex beams. We test the VCD measurement with two different systems: i) an experimental set of single circular nano-apertures drilled in a gold film with diameters ranging from 150 to 1950 nm; and ii) a theoretical set of golden spheres with the same diameters as the nano-apertures.

Results

We observe that in both systems, VCD > 0 for smaller diameters, VCD $\lesssim 0$ for intermediate values and VCD ≈ 0 for larger values of the diameter. Furthermore, the simulations show that a diffraction process characterized by a VCD ≈ 0 (VCD $\neq 0$) is polarization-independent (polarization-dependent). As a result, we relate VCD $\neq 0$ to a vectorial

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diffraction, and VCD ≈ 0 to a scalar one.

Conclusions

Overall, our results show compelling evidence that it is possible to experimentally assess the scalar/vectorial regime of a diffraction process, and that the VCD technique possesses the required features to measure the vectoriality of diffraction processes involving plasmonic cylindrically symmetric structures.

Keywords
Angular momentum, Vortex beam, Circular Dichroism, Helicity, Non-paraxial, Mie Theory, q-plate, Nanoholes



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REVISED **Amendments from Version 1**

In the revised version:

- There is a discussion about why the sign of VCD is positive for some diameters, and it is negative for some others.

- We mention that some other authors treat the problem using the framework of spin-orbit interaction.

- We comment on the role of the thickness of gold.

- Different sentences have been changed to make sure that this point is clear: VCD is not a universal measurement of diffraction vectoriality, but rather a technique that gives us a measure of vectoriality for our studied system.

- We have included a discussion on the differences between the CD and VCD measurements.

Any further responses from the reviewers can be found at the end of the article

Introduction

Light is an electromagnetic (EM) field that oscillates in the 1 THz – 30 PHz band (or 10 nm –300 μm in wavelength units)[1]. For classical phenomena, the propagation of light as well as its interaction with matter is described by Maxwell equations, which are typically expressed as four differential equations involving the electric and magnetic vector fields, $\{\mathbf{E}, \mathbf{B}\}$[2]. The six scalar functions $\{E_{1,2,3}, B_{1,2,3}\}$ that make up the EM field are not independent, as they are bonded to each other via Maxwell equations. That is, given a certain E_1 $(\mathbf{r}, t)$, the other two components $E_{2,3}$ $(\mathbf{r}, t)$ must have a certain functional form. Simply put, the spatial properties of light or the three intensity patterns given by E_i $(\mathbf{r}, t)$ cannot be independently modified. Or in other words, the polarization (or vectorial properties) of light cannot be modified without changing its spatial properties.

In optics, many phenomena are properly described using the wave optics approximation, *e.g.* interference[1], scalar diffraction theory[3] or holography[4]. In such approximation, the EM field of light is described as a scalar field times a polarization vector: $\mathbf{E}(\mathbf{r}, t) = E(\mathbf{r}, t)\,\mathbf{u}$, with $\mathbf{u}$ being a unit polarization vector. As a result, the spatial properties of the field (described by the scalar function $E(\mathbf{r}, t)$) and its polarization $\mathbf{u}$ can be independently modified. In particular, when a beam of light is described using wave optics, we typically say that we are in the paraxial approximation[5]. In contrast, we say that a beam is non-paraxial when its description requires a vectorial formulation[6]. Note that when it comes to light diffraction by matter, the description regime of this interaction does not necessarily need to be the same one as the beam. For example, the diffraction of a paraxial beam by a subwavelength spherical particle generally calls for a vectorial description, which is known as Mie Theory[7,8]. In the same way, the far-field diffraction of a non-paraxial beam off a macroscopic mirror could be described using scalar diffraction theory. Obviously, using the scalar (wave optics) approximation makes calculations as well as the interpretations of results much simpler, yet we need to make sure that we are under the right conditions to apply it. On the one hand, the scalar regime is suitable to describe diffraction processes where *either* the beam or the matter size are much greater than the wavelength. On the other hand, a vectorial description is needed when the dimensions of both matter and light beams are smaller than the wavelength. In contrast, the intermediate cases (which are common in nano-optics) are much more subtle, and it is difficult to know *a priori* if the scalar approximation can properly describe the interaction. This is schematically displayed in Figure 1, where we show the regimes of beam waist, wavelength and the matter dimension that roughly define the scalar approximation, the intermediate cases, and the purely vectorial diffraction regime.

Figuring out the diffraction regime has no secrets from a theoretical perspective, as the brute force approach could always be used. That is, any diffraction process could be modeled with both Maxwell equations and wave optics, and the results could be compared. If the difference between the diffracted EM fields computed in both ways was negligible for all points in space and time, we would conclude that the system behaves in a scalar way. Following the same logic, if the difference was not negligible, we would say that the diffraction process is vectorial. The rationale that is implicit in these two statements is that the difference between the EM field (in all points of space and time) computed using Maxwell equations and a wave optics approximation is a good *measure of vectoriality*. Obviously, this measure of vectoriality precludes its experimental realization, as it is impossible to measure the EM field in all points of space and time. In this work, we propose an experimental technique that can be used to define a measure of vectoriality which is zero when the diffraction process is scalar and different from zero when it is vectorial. We call this experimental technique Vortex Circular Dichroism (VCD).

VCD is a technique based on a Circular Dichorism (CD) measurement with vortex beams[9], i.e. beams that carry a phase singularity of the kind $e^{il\phi}$. The CD measurement is obtained as the differential absorption of the left and right circularly polarized (LCP and RCP) components of a light beam. CD is widely used in molecular and protein spectroscopy[10], among others. One of its most typical uses is the study of chiral properties of molecules[10]. It is generally accepted that the CD signal of a non-chiral object is zero, while the CD signal of a chiral object is different from zero. However, in recent years it has been demonstrated that vortex beams can induce a CD signal different from zero on non-chiral samples[11]. In this work[11], it was shown that there are two main differences between a standard CD measurement using Gaussian beams or plane waves, and the CD measurement using vortex beams: i) The CD measurement is a differential measurement between two beams that are connected via a mirror symmetry. That is, a LCP Gaussian beam and a RCP Gaussian beam are mirror images with respect to each other. In contrast, the VCD measurement uses a LCP and a RCP vortex beam, and it can be demonstrated that these two beams are not mirror symmetric images - the spiral phase of the vortex beam breaks this symmetry. ii) The two Gaussian beams (LCP and RCP) used in the CD measurement have an angular momentum content of 1 unit, whereas the angular momentum content of the two vortex beams used for

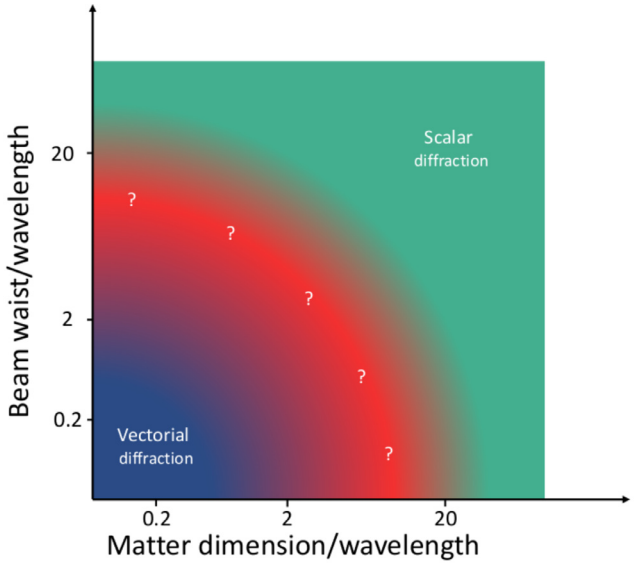

Figure 1. Sketch of the validity of the scalar diffraction approximation as a function of the beam waist, wavelength and matter dimensions. The image is only an indicative sketch, no calculations have been made to trace the limits between the different regions. Note that the vectorial diffraction theory is always valid, including the region where the scalar approximation can be used.

the VCD experiment have different angular momentum content equal to 0 and 2 units. Both i) and ii) are symmetry considerations, and they show why CD must be zero under normal incidence, and why VCD does not need to be zero. Yet, these symmetries consideration do not explain the physical mechanism that creates a non-zero VCD measurement for certain samples. Here, we shall prove that the underlying reason why a CD measurement using vortex beams (*i.e.* VCD) can yield a result different from zero with a non-chiral sample is the fact that the interaction between the vortex beam and the sample is vectorial. In other words, we will show that it is possible to experimentally assess if a diffraction process is scalar or vectorial, and that VCD is a measurement that has the right features to accomplish that.

To prove that, we have carried out a set of VCD measurements with cylindrical nanoholes (NH) with diameters ranging from 150 to 1950 nm (0.2 to 3.1 in diameter/wavelength units). Both the LCP and RCP vortex beams that are used for the VCD measurement have a phase singularity of order $l = -1$. Note that in general, vortex beams carrying angular momentum and beams with phase singularities are not equivalent[12,13]. But in this work, we use the terms vortex beam and beam carrying a phase singularity interchangeably because we use vortex beams with a single phase singularity of the kind $e^{il\phi}$. After the vortex beam has been created, the beams have been focused onto the NH with a microscope objective with an NA = 0.9 (MO_1), and they have been collected with another microscope objective with an NA = 0.8 (MO_2, see Figure 2). We have observed that the smaller NHs yield VCD signals which are much greater than zero, whereas the larger ones yield a VCD which is approximately zero. Our main claim is that VCD is assessing the vectoriality of this diffraction process, yielding signals similar to zero when the interaction is scalar and signals different from zero when the interaction is vectorial. In order to gain a better understanding of the VCD measurement and confirm our hypothesis, we have also performed some numerical simulations for a different system which shares all the symmetries of our experimental samples but it is easier to simulate: a spherical particle. Using analytical Mie Theory for non-paraxial beams[8,14], we have computed the theoretical VCD for spheres with the same diameters as the NHs used in the experiment. Not only we have obtained very good qualitative agreement with the experimental data, but also we have been able to verify that the VCD signal is related to the degree of polarization-dependence in the diffraction process. We have found that VCD $\neq 0$ (VCD ≈ 0) indicates that the diffraction process is polarization-dependent (independent). Overall, our results show that the vectoriality of a diffraction process can be experimentally assessed, and that VCD provides a good measure of the scalar/vectorial regime of a diffraction process for plasmonic cylindrically symmetric systems.

Methods

Optical measurements

The optical set-up in order to perform the VCD measurements is schematically shown in Figure 2. A single mode fiber-coupled HeNe laser (λ = 632.8 nm; Thorlabs HNL210LB) creates a Gaussian beam. The beam goes through a power control system made of a set of neutral density filters (NDF; Thorlabs NDC-100C-2M), a half-wave plate (HWP; Optique J. Fichou, LO-633) and a linear polarizer (LP; Thorlabs GTH10M-A). Then, the polarization is changed to RCP with a quarter-wave plate (QWP: Optique J. Fichou, LO-633). The RCP Gaussian beam then goes through a q-plate[15] of order $q = 1/2$, which turns the RCP to LCP and adds a spiral phase of the kind $e^{-i\varphi}$. After the q-plate, we eventually place another HWP. When inserted, this HWP changes the handedness of the state from LCP to RCP. In both cases the setup produces a circularly polarized vortex beam with a phase singularity of order $l = -1$. Then, the

vortex beam hits the back-aperture of a microscope objective MO_1 (Olympus, MPLANFL N 100X NA0.9), which has an NA = 0.9. It is important to note here due to the focusing of MO_1, the polarization at the NH plane (focal plane) is not circularly polarized (see Figure 5). The circular NH sits on top of a sample holder which is attached to a position system composed of a micro and a nanopositioner (PI P-562.3CD). The NHs that are used in this experiment range from 150 nm to 1950 nm, in steps of 100 nm. This range corresponds to 0.2 - 3.1 in diameter/wavelength units. For the smaller NHs, positioning steps of the order of 5 nm need to be taken in order to make sure that the alignment is correct, and that the cylindrical symmetry of the system formed by the sample and beam is maintained. The transmission of the beam through the NH is collected with MO_2 (Olympus LMPLANFL 100X NA0.8), in this case with an NA = 0.8. The NH lies at the focal plane of MO_2, therefore the transmission is collimated. The collimated transmission goes through a lens of f = 150 mm, which images the transmitted light at the NH plane onto a CMOS camera (The imaging source DMK33UX290). The CMOS camera allows us to measure the differential transmission of the LCP and RCP vortex beams. In order to get a good signal-to-noise ratio for the VCD measurements, we adjust the power of the laser so that the power at the CMOS chip of the camera is at about 80% of the saturation limit. For each of the NH sizes in the 150 - 1950 nm range, we have probed four independent single NHs. All four NHs have been fabricated with the same target diameter. The VCD measurement of each of the four single NHs has been statistically obtained as the result of 80 (2×2×20) snapshots. First, we align the NH with respect to the LCP/RCP vortex beam and take 40 snapshots (2×20) to measure its transmission $I_l^{x,t}$, with x = *LCP/RCP*. In order to obtain the $I_l^{x,t}$ number, we isolate the area where the beam signal is located in each snapshot, we add the counts from that area, and we subtract the noise signal. This yields $I_{l,i}^{x,t}$. Then, $I_l^{x,t}$ is obtained as the average of the 20 $I_{l,i}^{x,t}$ values. Immediately after, we move the sample at 10 μm of distance and take 40 (2×20) snapshots to measure the transmission of the LCP/RCP through the gold film $I_l^{x,in}$. We compute $I_l^{x,in}$ with the same protocol that we use to compute $I_l^{x,t}$. Then, we compute the normalized transmission $I_l^x = I_l^{x,t} / I_l^{x,in}$, and finally $\mathrm{VCD}_l = I_l^{LCP} - \mathrm{IlRCP}$. We are mostly interested in the VCD trend-line as a function of the NHs diameters, so we divide the VCD_l result over 10000 so that the result is of the order of 1. Our quick normalization method allows us to correct the power oscillations of the fibre-coupled laser.

Nanofabrication

First, we have deposited a 200 nm gold (Au) layer on top of a 150 μm cover glass. An adhesion layer of 5 nm of titanium (Ti) has been added in between the gold and glass. The deposition of the Au and Ti layers have been carried out by sputtering deposition. Then, the sample composed of the Au, Ti, and Glass layers has been nano-patterned by means of a focused ion beam (FIB). Using the FIB, we have managed to fabricate single circular NHs with diameter sizes ranging from 150 to 1950 nm. Figure 3 shows some of the NHs that have been fabricated. We have fabricated four NHs for each of the target sizes. We have left a distance of at least 25μm between each NH and its closer neighbors in order to avoid any array effects. Note that the thickness of the Au layer has been chosen to greatly reduce the direct transmission of the beams through the sample. It is not expected that slight changes in the thickness of the gold layer will modify the results of this paper.

Numerical simulations

All the numerical simulations included in this manuscript are carried out in the framework of analytical Mie Theory[16]. The code used to run the simulations is semi-analytical and it is scripted in Matlab. The code solves the classical Mie Theory equations for a non-paraxial vortex beam excitation. An open-source solver for Mie Theory is available on GitHub here. The non-paraxial excitation can be modelled following the equations in ref. 14. The code takes an incident circularly polarized

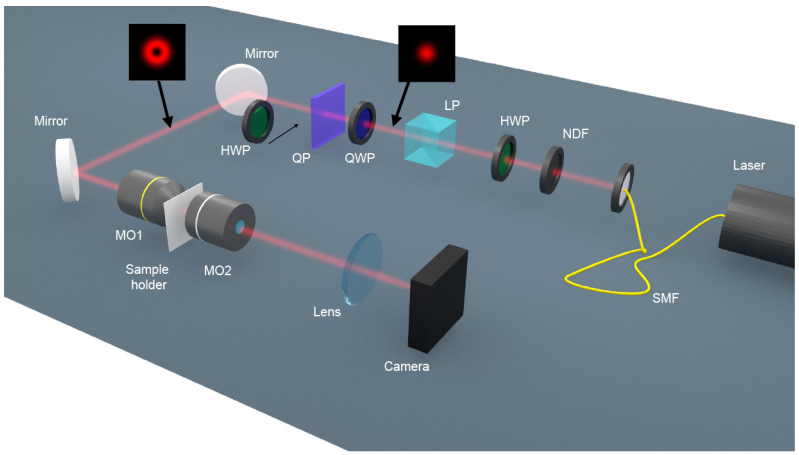

Figure 2. Schematics of the optical set-up that has been used to measure the Vortex Circular Dichroism signal from the cylindrical nanoholes. SMF: single mode fiber. NDF: neutral density filter. HWP: half-wave plate. LP: linear polarizer. QWP: quarter-wave plate. QP: q-plate. MO: microscope objective.

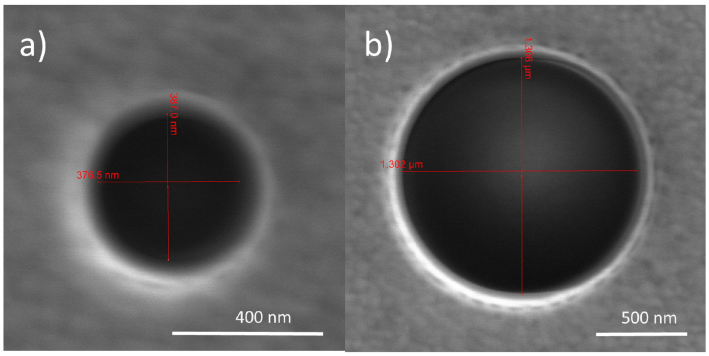

Figure 3. Scanning electron microscope (SEM) images of two nanoholes (NHs), with a diameter of **a**) 380 nm and **b**) 1305 nm.

paraxial Laguerre-Gaussian beam[17]; it focuses it using the aplanatic lens model[18] with an NA= 0.9; it decomposes the beam into the multipolar basis[19]; and then it computes the interaction between this incoming beam and the sphere. As a result, the EM field outside and inside of the sphere is obtained, as well as the scattering and absorption cross sections[14,20]. The theoretical VCD is computed by subtracting the absorption cross sections of the LCP and RCP vortex beams used in the experiment. Note that our absorption cross sections σ_{abs} are dimensionless, and they are computed as the ratio between the absorbed power over the incident power of the beam, which is normalized to 1. Then, the arbitrary units of VCD used in Figure 4b) are the dimensionless cross section units multiplied by a factor of 1000 to make them be of the order of 1. Besides, the scattered EM field is expressed in the basis of $\{\hat{\mathbf{e}}_+, \hat{\mathbf{e}}_-, \hat{\mathbf{z}}\}$, where $\hat{\mathbf{e}}_\pm = \left(\hat{\mathbf{x}} \mp i\hat{\mathbf{y}}\right)/\sqrt{2}$. Expressing the field in this basis allows us to easily track changes in the polarization of the incident beam.

Results

The results of our experiments can be observed in Figure 4a), and the full data is available from *Underlying data*[21]. Figure 4a) shows the variation of VCD as a function of the diameter of the NHs. Notice that describing the VCD measurement as the differential absorption of LCP and RCP vortex beams is an abuse of language, as the two vortex beams are tightly focused, and their polarization at the NH focal plane is not circular (see Figure 5). However, we can use these terms when referring to the polarization state at the rear-aperture of MO_1. Thus, for the sake of simplicity, we have chosen to extend this notation to the focusing region. It is also important to note that the VCD measurement is performed as a differential power measurement. Because of that, it is of paramount importance to normalize the transmission to the incident power:

$$\mathrm{VCD}_l = I_l^{LCP} - I_l^{RCP} \tag{1}$$

where $I_l^{LCP}\left(I_l^{RCP}\right)$ is the normalized transmitted power of a LCP (RCP) vortex beam with a phase singularity of order l. That is, $I_l^x = I_l^{x,t} / I_l^{x,in}$ with x = *LC P/RCP*, where $I_l^{x,t}$ is the measured transmitted power of LCP/RCP beams through the NHs, and $I_l^{x,in}$ is proportional to the value of the power of the incident LCP/RCP beams (see Methods). In our experiment, we have used $l = -1$. As a result, the total angular momentum of the two beams that are used for the VCD measurement is 0 (LCP) and -2 (RCP)[9,17,22,23]. We have chosen $|l| = 1$ because vortex beams with phase singularities of higher orders are very unstable and they tend to split[24,25], thus making the VCD measurement less reproducible. The sign of l has been chosen so that the VCD measurement for small NH sizes was positive. Note that the sign of l has a clear impact on the sign of the VCD, as it can be proven that $\mathrm{VCD}_l = -\mathrm{VCD}_{-l}$[11]. We have carried out VCD measurements for single NHs whose sizes range from 150 to 1950 nm in steps of 100 nm. In diameter/wavelength units this corresponds to 0.2 to 3.1 in size. For each size, we have probed four single NHs. As a result, in Figure 4a), we have plotted four independent data points for each size. Each VCD_l data point is obtained as a function of 80 snapshots see Methods). The error bars of each individual point in Figure 4a) are not seen as they are smaller than the markers themselves. Moreover, we have also plotted two trend-lines that follow the minima and maxima VCD values for each diameter size to help the eye of the reader. Now, it can be observed that the experimental data shown in Figure 4a) can be split into three different areas. When the diameter of the NHs is approximately below 700 nm (1.1 in diameter/wavelength units), the VCD signal is positive. When the diameter is approximately in between 700 and 1600 nm, VCD yields a negative (but closer to zero) signal. And for diameter sizes greater than 1600 nm (2.5 in diameter/wavelength units), the VCD signal is approximately zero. Figure 4a) is the main experimental result of this work, and it shows that the polarization of a first order non-paraxial vortex beam plays a significant role in the transmission of light through NHs when the size of the NH is below 1600 nm (or 2.5 in diameter/wavelength units). For greater sizes, the polarization does not play any role. That is, the diffraction of our non-paraxial beam through a NH is scalar for sizes above 1600 nm, and it is vectorial for sizes below 700 nm. In between there is an intermediate regime where diffraction is generally vectorial, but it is not far from being scalar - as shown by the crossings of the VCD trend-line with the line VCD = 0. Figure 4a) corroborates what had been sketched in Figure 1: delimiting the vectorial/scalar interaction regime is a difficult endeavor which requires a calculation

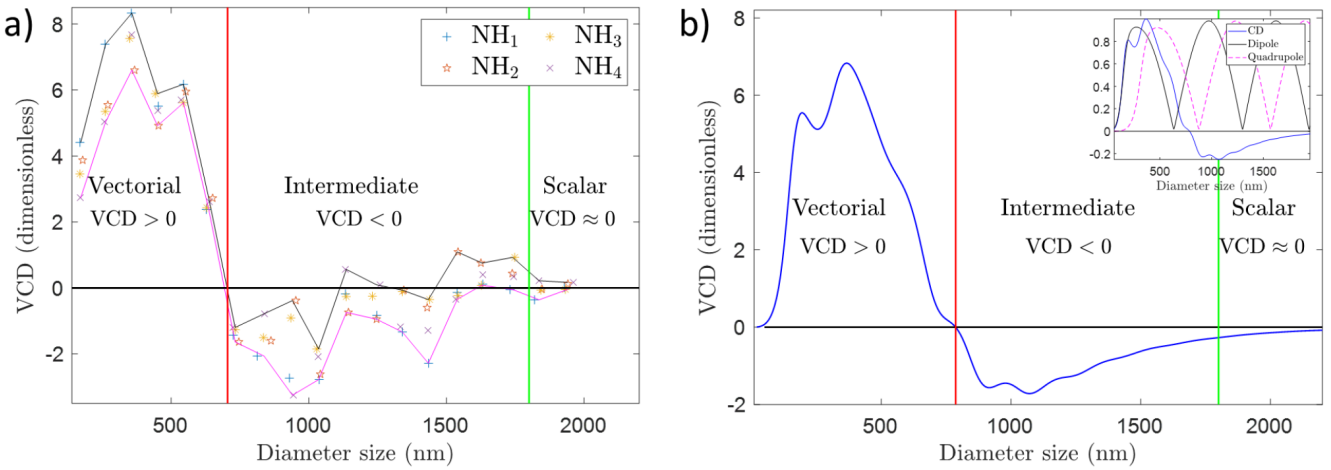

Figure 4. (**a**) Vortex Circular Dichroism (VCD) measurements for a set of nanoholes (NHs) with sizes ranging from 150 to 1950 nm, which correspond to a diameter/wavelength ratio ranging from 0.2 - 3.1 approximately. The laser operates at 632.8 nm. The VCD measurement is defined as the normalized differential transmitted intensity for two vortex beams with opposite circular polarization. The vortex beam that has been used has a phase singularity of order $l = -1$, and it has been focused using a microscope objective with NA= 0.9. The dimensionless units that account for VCD changes are normalized power units (see Methods). (**b**) Theoretical VCD as a function of the diameter size of a gold particle. The diameter range corresponds to a diameter/wavelength ratio ranging from 0.2 - 3.1 approximately. VCD is computed as $\mathrm{VCD} = \sigma^{+}_{abs} - \sigma^{-}_{abs}$. The intensity profiles of the incoming beams are given in Figure 5. The wavelength is 632.8 nm. The vortex beam has $l = -1$ and it has been focused with an aplanatic lens with NA= 0.9. The dimensionless units of VCD are explained in Methods. The inset shows the theoretical VCD signal and the dipolar and quadrupolar resonances of the sphere. None of the two resonances peaks at the same position as the VCD signal.

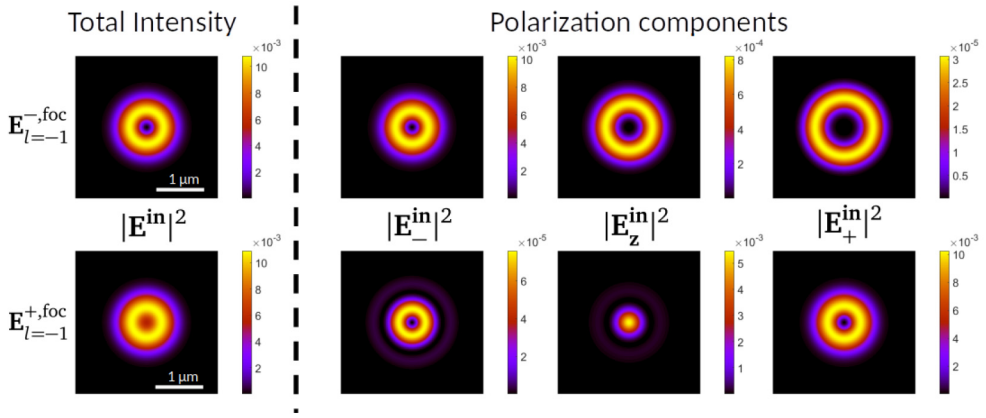

Figure 5. On the left of the dashed vertical line, modulus square of the electric field of the incoming beams $\mathbf{E}^{-,\mathrm{foc}}_{l=-1}$ (top) and $\mathbf{E}^{\pm,\mathrm{foc}}_{l=-1}$ (bottom). On the right, projections in the three polarization components $\{\hat{\mathbf{e}}_{-}, \hat{\mathbf{z}}, \hat{\mathbf{e}}_{+}\}$ of $\mathbf{E}^{-,\mathrm{foc}}_{l=-1}$ (top) and $\mathbf{E}^{\pm,\mathrm{foc}}_{l=-1}$ (bottom). All the plots are taken at a transverse 4 × 4 μm^2 XY plane at the focal plane of the incoming beam. The colorbars show that the differences in intensity between the three polarization components vary across two orders of magnitude. Both $\mathbf{E}^{\pm,\mathrm{foc}}_{l=-1}$ have been obtained applying the aplanatic lens model with an NA=0.9 to a paraxial vortex beam given by equation (2) for λ = 632.8 nm.

or a measurement. Our results demonstrate that VCD is a good candidate to quantify the scalar/vectorial regime, since i) it consistently defines the trivial vectorial boundary for NH sizes clearly smaller than λ; ii) it captures the subtle vectorial behavior of diffraction in the intermediate cases; iii) it trends towards a scalar behavior for NH sizes larger than λ; iv) and it shows a good qualitative agreement with the theoretical VCD reported in Figure 4b). Interestingly, Figure 4a) also shows us that the maximum VCD signal does not happen for the smallest NHs, but rather for some intermediate sizes. Using Mie Theory, we have checked that this is a rather convoluted effect, and it is not directly linked to a single internal resonance of the structure. This can be observed in the inset of Figure 4b), where we have plotted the theoretical VCD signal as well as the dipolar and quadrupolar resonances of a golden sphere. It is seen that none of the two resonances has a maximum at sizes that maximize the VCD signal.

Discussion

In this section, we carry out some numerical simulations that will help us to strengthen our claims on the validity of VCD as a quantitative technique to determine the scalar/vectorial

nature of diffraction. First of all, we have simulated the two focused incoming beams that we use in the VCD measurement, *i.e.* LCP and RCP focused vortex beams with $l = -1$. The analytical expression of their electric field at the rear aperture of MO_1 can be expressed as:

$$\mathbf{E}^{\pm}_{l=-1} = A\rho^{-1}L_0^{-1}\left(\frac{2\rho^2}{w_0^2}\right)e^{-\rho^2/w_0^2}e^{i(-\phi+kz)}\hat{\mathbf{e}}_{\pm} \tag{2}$$

where A is a normalization constant; $\{\rho, \varphi, z\}$ are the cylindrical coordinates, with z being the propagation axis of the beam; L_0^{-1} is a Laguerre polynomial of order $l = -1$; w_0 is the beam waist of the beam at the rear aperture of MO_1; $k = 2\pi/\lambda$ is the wavevector, and $\hat{\mathbf{e}}_{\pm}$ the polarization vector for LCP (RCP) depending on the + (−) sign. Note that the ± sign has to do with the handedness of light, or its helicity[9,22]. In order to obtain the electric field at the focal plane of MO_1, we use the paraxial beam in equation (2) $\mathbf{E}^{\pm}_{l=-1}$ and apply the aplanatic lens model[18], yielding $\mathbf{E}^{\pm,\text{foc}}_{l=-1}$. In Figure 5, we display $|\mathbf{E}^{\pm,\text{foc}}_{l=-1}|^2$, that is, the modulus square (intensity) of the electric field distribution of $\mathbf{E}^{\pm}_{l=-1}$ at the focal plane of a lens with NA = 0.9 for a λ = 632.8 nm. The top and bottom rows display the intensity of the $\mathbf{E}^{-,\text{foc}}_{l=-1}$ and $\mathbf{E}^{+,\text{foc}}_{l=-1}$ beams respectively. Each row is composed of four images, corresponding to the total intensity, and its projections into the three polarization components $\hat{\mathbf{e}}_-$, $\hat{\mathbf{e}}_z$, $\hat{\mathbf{e}}_+$. We observe that the two beams have a different intensity profile, where $\mathbf{E}^{+,\text{foc}}_{l=-1}$ concentrates more energy around the center of symmetry due to the Gaussian-like $\hat{\mathbf{z}}$ component[9,11,22]. Now, given these two incoming focused vortex beams, we simulate the VCD that they induce on spherical golden particles. To do that, we decompose the incoming beams into multipoles using the method put forward in Refs. 14,17 and solve the analytical Mie Theory problem. As a result, we obtain the scattered EM field $\mathbf{E}^s$ as well as the scattering and absorption cross sections[16]. We compute the theoretical VCD as:

$$\text{VCD} = \sigma^{+}_{abs} - \sigma^{-}_{abs}. \tag{3}$$

where both $\sigma^{\pm}_{abs}$ are the absorption cross sections obtained with a focused vortex beam with $l = -1$, and where the +(−) sign has to do with a LCP (RCP) before the back-aperture of the focusing lens. Notice that our theoretical modelling is based on Mie Theory and a beam description based on the total angular momentum and handedness, yet some other authors have addressed this problem in the framework of spin-orbit interactions[26–28].

In Figure 4b), we plot the theoretical VCD obtained with the two beams plotted in Figure 5 as a function of the sphere diameter. Note that we normalize the plot in a way such that the values that we obtain are in the order of 1 (see Methods). We observe that we recover most of the features of Figure 4a). We see that the VCD yields a positive result for the smaller diameters. We also observe that there is an intermediate zone where VCD yields negative (yet closer to zero) values. The scalar zone of VCD ≈ 0 for greater particle sizes is also present in Figure 4b). Besides, we also see that the relative variations in VCD values (in between -2 and 6 given our normalization, see Methods) are comparable to those observed in Figure 4a), despite the fact that Figure 4b) is carried out for a single sphere, which clearly is a different system from a single NH despite sharing some symmetries (mirror and cylindrical). Finally, notice that changes in the geometry, material or NA, would certainly have an effect in the experimental and theoretical VCD trend line. Yet, the comparison between Figures 4a) and 4b) still leads us to believe that VCD has a good potential to probe the vectoriality of diffraction processes for plasmonic cylindrically symmetric systems.

In order to further support this claim, in Figure 6 we plot the modulus square of the scattered field off the sphere at a transverse XY plane, at a distance $z = 2$ μm from the center of the sphere. We use $\mathbf{E}^{+,\text{foc}}_{l=-1}$ as the incoming beam, which is plotted as the top row in Figure 6. A scalar diffraction process is characterized by a negligible role of the polarization of light. In other words, a scalar diffraction process will not change the polarization state of an incoming beam, because the interaction is polarization-independent. The bottom row of Figure 6 shows exactly that: the intensity pattern as well as the polarization projections of the scattered field off a sphere of 2000 nm of diameter are almost the same as the incident beam $\mathbf{E}^{+,\text{foc}}_{l=-1}$ (see Figure 6, top row). That is, most of the intensity comes from the $\hat{\mathbf{e}}_+$ polarization component, whereas the $\{\hat{\mathbf{e}}_-, \hat{\mathbf{z}}\}$ components are negligible. And indeed, this corresponds to a case of VCD ≈ 0. In contrast, the middle row of Figure 6 shows exactly the opposite, *i.e.* a sphere with a diameter of 200 nm strongly changes the polarization of the incident beam. In this case, indeed the scattered field is not dominated by a single polarization component, instead the three $\{\hat{\mathbf{e}}_-, \hat{\mathbf{z}}, \hat{\mathbf{e}}_+\}$ polarization components yield a similar intensity (see the colorbars in Figure 6). This is a consequence of the fact that the interaction is vectorial, and therefore it is not polarization-independent. And as expected, VCD ≠ 0.

That is, the simulations presented in Figure 6 support the main claim of this article, namely that VCD is an experimental technique that allow us to assess the scalar/vectorial regime of a diffraction process.

Conclusions

Vortex Circular Dichroism (VCD) is an experimental technique that consists in carrying out a circular dichroism (CD) measurement with a vortex beam. We have presented some experimental data and theoretical simulations to support the following three claims. i) The vectoriality of a diffraction process can be experimentally assessed. ii) The transition between the scalar and vectorial regimes in a diffraction process is subtle and not easy to predict, hence its characterization calls for a measuring technique. iii) VCD is a potential candidate to assess the scalar/vectorial regime of a diffraction process. The experimental data has been collected for single nanoholes in a gold film, while the theoretical calculations have been done for golden spheres. Despite having different geometries, both systems have yielded a very similar trend-line of VCD vs size of the nanostructure (see Figure 4): VCD > 0 for small sizes, VCD $\lesssim$ 0 for intermediate sizes and VCD ≈ 0 for larger sizes. Some extra simulations of the scattered far-field (see Figure 6) have corroborated that scalar (vectorial) diffraction processes in

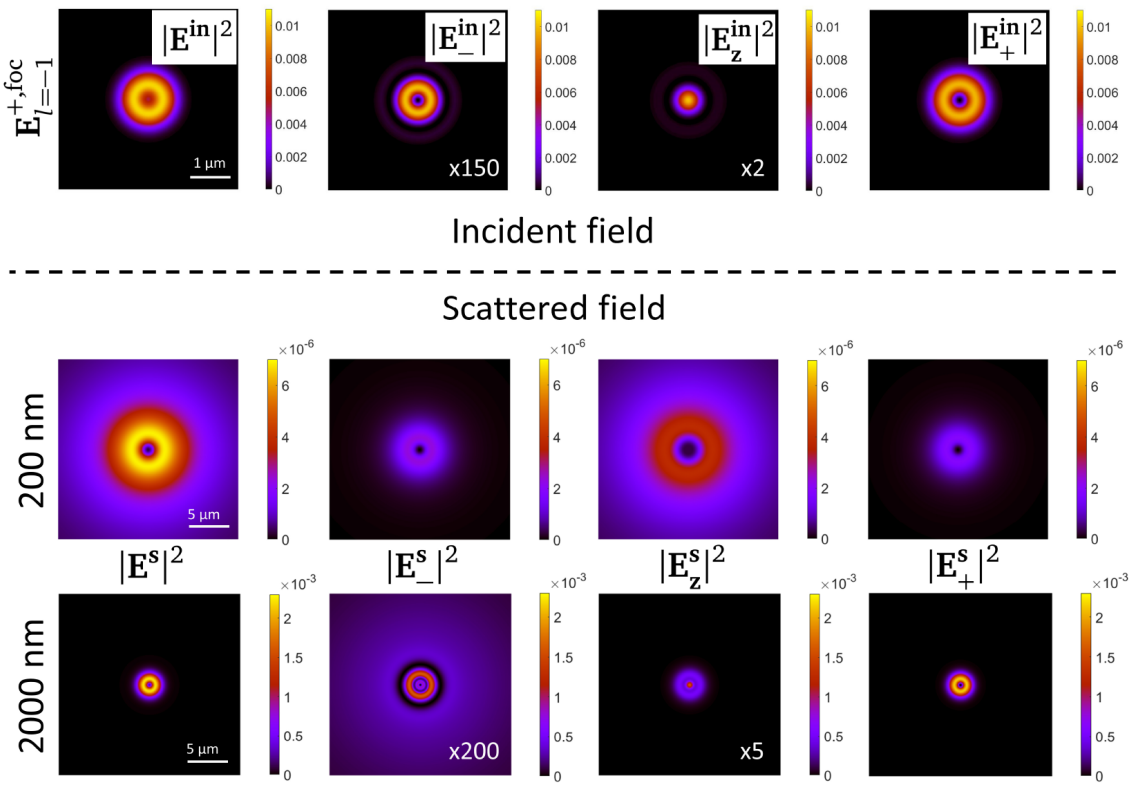

Figure 6. Modulus square of the scattered field $\mathbf{E}^s$ as well as its projection in the three polarization components $\{\hat{\mathbf{e}}_-,\hat{\mathbf{z}},\hat{\mathbf{e}}_+\}$ for two spheres with diameters equal to 200 nm (middle row) and 2000 nm (bottom row). The incident field inducing the scattered field is $\mathbf{E}^{+,foc}_{l=-1}$, and its intensity profile as well as its polarization components at the focal plane are plotted in the top row of the image. The scattering plots are taken at a transverse 40 × 40 μm^2 XY plane at distance of $z = 2$ μm of the center of the sphere. The colorbars of all the plots in one row are the same. The intensity of some of the polarization components has been multiplied times a factor. The factor has been written in the plot as ×5.

which VCD ≈ 0 (VCD ≠ 0) are (not) polarization-independent. In our experiment we considered a diffraction process involving a vortex beam with a phase singularity of the kind $e^{il\phi}$. Note that if the measurement was done with a Gaussian beam, no information about the scalar/vectorial regime would be found, as the CD measurement would be 0 for all the sizes. This is due to the fact that a LCP and RCP Gaussian beam are mirror symmetric, whereas the LCP and RCP vortex beams used in VCD are not[9,11]. Overall, our work shows that VCD has all the necessary features to be a measure of vectoriality of a diffraction process for plasmonic cylindrically symmetric matter structures. It is possible that another possible measure to assess the scalar/vectorial regime could involve other non mirror-symmetric beam pairs such as, for instance, the well known radial and azimuthal polarized beams[29,30]. As it happens with most of new measuring techniques, its direct applications are still to be thought of. Yet it is clear that the experimental quantification of the scalar/vectorial regime could contribute to gain a more in-depth knowledge of fundamental light-matter interactions. In particular, it could be an important asset in order to program experiments and/or simulations in an optimized way, *e.g.* saving a vectorial simulation and running a scalar one instead.

Ethics and consent

Ethical approval and consent were not required.

Data availability

Underlying data

Zenodo: Vortex Circular Dichroism experiment at IIT. https://doi.org/10.5281/zenodo.7319991[21].

This project contains the following underlying data, sorted in three different files:

- *Snapshots.zip*. This compressed file contains all the snapshots that have been taken in order to carry out the VCD measurements. As stated in *Methods*, there are four categories of snapshots concerning the transmission of vortex beams under four different conditions: 1) Transmission of LCP vortex beams through the nanohole; 2) Transmission of RCP vortex beams through the nanohole; 3) Transmission of LCP vortex beams through the gold layer; 4) Transmission of RCP vortex beams through the gold layer. The notation of the files is `(n)h[size in nm]_[pol]_[pos]_[num].tiff`, where `h (nh)` is written when the light passes (does not pass) through

the nanohole. `[size in nm]` gives us the diameter of the nanohole in nm. `[pol]` tells us if the incident beam is LCP (`[pol]=60`) or RCP (`[pol]=0`). `[pos]=1,2,3,4` refers to the four nanoholes that have been probed for each diameter size. `[num]` is the snapshot number, as 10 snapshots were taken for each nanohole.

- *Data.ods*. This file contains two sets of data, sorted in two different tabs. In the *Conversion Table* tab, we find a table that allows us to give units to the snapshots given in the *Snapshots.zip* file. The table is needed because in the measurements we regulated the power of the laser to almost saturate the CMOS chip of the camera. Moreover, the same table can be later used to turn the VCD measurement dimensionless. Then, in the *Nanoholes sizes* tab, we use the snapshots collected in the *SEM_nanoholes.zip* file to retrieve the sizes of the nanoholes that have been used in the experiment.
- *SEM_nanoholes.zip*. This file contains all the SEM measurements (.jpeg) to retrieve the diameter of the nanoholes used in the experiment.

Data are available under the terms of the Creative Commons Attribution 4.0 International license (CC-BY 4.0).

Acknowledgements

The authors want to thank Carlo Forestiere for useful discussions. X.Z.-P. wants to thank Claudio Biagini and Fabio Moia for their valuable help in the fabrication of the nanoholes. X.Z.-P. also would like to acknowledge that the fabrication of the samples was done in the Clean Room facility of IIT.

References

1. Saleh BEA, Teich MC: **Fundamentals of photonics**. Wiley New York, 1991; **22**. .
Publisher Full Text
2. Jackson JD: **Classical electrodynamics: Third Edition**. *Am J Phys.* John Wiley & Sons, New York, 1999; **67**. (9): 841.
Publisher Full Text
3. Gupta SD, Ghosh N, Banerjee A: **Wave optics: basic concepts and contemporary trends**. CRC sress, 2015.
Publisher Full Text
4. Sirohi RS: **Wave optics and its applications**. Orient Blackswan, 1993.
Reference Source
5. Lax M, Louisell WH, McKnight WB: **From maxwell to paraxial wave optics.** *Phys Rev A.* 1975; **11**(4): 1365.
Publisher Full Text
6. Vaveliuk P, Ruiz B, Lencina A: **Limits of the paraxial approximation in laser beams.** *Opt Lett.* 2007; **32**(8): 927–929.
PubMed Abstract | Publisher Full Text
7. Mie G: **Beiträge zur optik trüber medien, speziell kolloidaler metallösungen.** *Annalen der Physik.* 1908; **330**(3): 377–445.
Publisher Full Text
8. Gouesbet G, Gréhan G: **Generalized lorenz-mie theories**. Springer, 2011.
Publisher Full Text
9. Zambrana-Puyalto X, Vidal X, Fernandez-Corbaton I, *et al.*: **Far-field measurements of vortex beams interacting with nanoholes.** *Sci Rep.* 2016; **6**: 22185.
PubMed Abstract | Publisher Full Text | Free Full Text
10. Barron LD: **Molecular light scattering and optical activity**. Cambridge University Press, 2009.
Publisher Full Text
11. Zambrana-Puyalto X, Vidal X, Molina-Terriza G: **Angular momentum-induced circular dichroism in non-chiral nanostructures.** *Nat Commun.* 2014; **5**: 4922.
PubMed Abstract | Publisher Full Text
12. Berry MV: **Superoscillations and the quantum potential.** *Eur J Phys.* 2020; **42**(1): 015401.
Publisher Full Text
13. Berry MV: **The singularities of light: intensity, phase, polarisation.** *Light Sci Appl.* 2023; **12**(1): 238.
PubMed Abstract | Publisher Full Text | Free Full Text
14. Zambrana-Puyalto X, Vidal X, Molina-Terriza G: **Excitation of single multipolar modes with engineered cylindrically symmetric fields.** *Opt Express.* 2012; **20**(22): 24536–24544.
PubMed Abstract | Publisher Full Text
15. Marrucci L, Manzo C, Paparo D: **Optical spin-to-orbital angular momentum conversion in inhomogeneous anisotropic media.** *Phys Rev Lett.* 2006; **96**(16): 163905.
PubMed Abstract | Publisher Full Text
16. Bohren CF, Huffman DR: **Absorption and scattering of light by small particles**. Wiley, 1983.
Reference Source
17. Zambrana-Puyalto X: **Control and characterization of nano-structures with the symmetries of light**. PhD thesis, Macquarie University, 2014.
Reference Source
18. Novotny L, Hecht B: **Principles of nano-optics**. Cambridge University Press, 2006.
Publisher Full Text
19. Rose ME: **Elementary theory of angular momentum**. Wiley, New York, 1957.
Reference Source
20. Zambrana-Puyalto X, Molina-Terriza G: **The role of the angular momentum of light in mie scattering. excitation of dielectric spheres with laguerre-gaussian modes.** *J Quant Spectrosc Radiat Transf.* 2013; **126**: 50–55.
Publisher Full Text
21. Zambrana-Puyalto X: **Vortex circular dichroism experiment at IIT.** [Data set]. Zenodo. 2022.
http://www.doi.org/10.5281/zenodo.7319991
22. Fernandez-Corbaton I, Zambrana-Puyalto X, Molina-Terriza G: **Helicity and angular momen-tum: A symmetry-based framework for the study of light-matter interactions.** *Phys Rev A.* 2012; **86**: 042103.
Publisher Full Text
23. Zambrana-Puyalto X, Vidal X, Woźniak P, *et al.*: **Tailoring multipolar mie scattering with helicity and angular momentum.** *ACS Photonics.* 2018; **5**(7): 2936–2944.
Publisher Full Text
24. Ricci F, Löffler W, Van Exter MP: **Instability of higher-order optical vortices analyzed with a multi-pinhole interferometer.** *Opt Express.* 2012; **20**(20): 22961–22975.
PubMed Abstract | Publisher Full Text
25. Neo R, Tan SJ, Zambrana-Puyalto X, *et al.*: **Correcting vortex splitting in higher order vortex beams.** *Opt Express.* 2014; **22**(8): 9920–9931.
PubMed Abstract | Publisher Full Text
26. Bliokh KY, Alonso MA, Ostrovskaya EA, *et al.*: **Angular momenta and spin-orbit interaction of nonparaxial light in free space.** *Phys Rev A.* 2010; **82**: 063825.
Publisher Full Text
27. Bliokh KY, Rodríguez-Fortuño F, Franco N, *et al.*: **Spin–orbit interactions of light.** *Nature Photon.* 2015; **9**(12): 796–808.
Publisher Full Text
28. Vázquez-Lozano JE, Martínez A: **Classical emergence of intrinsic spin-orbit interaction of light at the nanoscale.** *Phys Rev A.* 2018; **97**(3): 033804.
Publisher Full Text
29. Banzer P, Kindler J, Quabis S, *et al.*: **Extraordinary transmission through a single coaxial aperture in a thin metal film.** *Opt Express.* 2010; **18**(10): 10896–10904.
PubMed Abstract | Publisher Full Text
30. Woźniak P, Banzer P, Leuchs G: **Selective switching of individual multipole resonances in single dielectric nanoparticles.** *Laser Photonics Rev.* 2015; **9**(2): 231–240.
Publisher Full Text

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Version 2

Reviewer Report 30 May 2024

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✔

Andrew Forbes
School of Physics, University of the Witwatersrand, Johannesburg, South Africa

No further comments to make.

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: structured light

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Version 1

Reviewer Report 12 May 2023

https://doi.org/10.21956/openreseurope.16119.r31133

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?

Andrew Forbes
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The authors question the onset of regimes where vectorial light treatment is needed and where scalar is ok. To do this they pass light through nanoholes of various sizes in relation to the wavelength and look at the effect on CD as a test. Everything looks solid except for the generalised

conclusions. Finding a trend for THIS particular system and something similar in one other system does not imply that it is universally true. As far as I could see, each measurement was performed only once, so I doubt one can make sweeping claims. Also, the difference between CD and VCD was buried in the text and could sure be brought our more (mathematically) since it is central to the story. On a minor note, the technical terms are often a little odd, e.g., unitary has a specific meaning and is not usually used to describe a unit vector, and singular beams with a phase singularity and vortex beams (carrying OAM) are not the same thing - see recent papers by Michael Berry on the topic.

Having said this, I do like the simplicity of the question and its experimental test, so just a few adjustments to the main claims or at least a discussion for the reader to consider arguments would be in order.

Is the work clearly and accurately presented and does it cite the current literature?
Yes

Is the study design appropriate and does the work have academic merit?
Yes

Are sufficient details of methods and analysis provided to allow replication by others?
Yes

If applicable, is the statistical analysis and its interpretation appropriate?
No

Are all the source data underlying the results available to ensure full reproducibility?
Yes

Are the conclusions drawn adequately supported by the results?
Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: structured light

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard, however I have significant reservations, as outlined above.

Author Response 29 Apr 2024

Xavier Zambrana-Puyalto

We thank Andrew Forbes for reviewing our article. His comments have definitely helped us to improve our work. Next, we address his points, one by one. To see a more complete response with figures, equations and references, see this PDF file attached here.

- We thank the referee for pointing this out. We tried to be careful, as we never wanted to imply that VCD is a universal measurement of the vectoriality of diffraction. We think that it is rather a technique that works for the cases that we have studied. Due to its features, we envision that its use could be extended to other cases, but that needs to be demonstrated, and this was not the aim of this work. As a result, we have changed several sentences in the manuscript to make this clear. For example, we have changed the last sentence of the abstract to: "Overall, our results show compelling evidence that it is possible to experimentally assess the scalar/vectorial regime of a diffraction process, and that the VCD technique possesses the required features to measure the vectoriality of diffraction processes involving plasmonic cylindrically symmetric structures."

We thank the referee for the comment about the differences between the CD and VCD measurements. It is indeed central to the story, and it needs to be highlighted. In the revised version, we have included the following sentences:

- "In this work [11], it was shown that there are two main differences between a standard CD measurement using Gaussian beams or plane waves, and the CD measurement using vortex beams: i) The CD measurement is a differential measurement between two beams that are connected via a mirror symmetry. That is, a LCP Gaussian beam and a RCP Gaussian beam are mirror images with respect to each other. In contrast, the VCD measurement uses a LCP and a RCP vortex beam, and it can be demonstrated that these two beams are not mirror symmetric images - the spiral phase of the vortex beam breaks this symmetry. ii) The two Gaussian beams (LCP and RCP) used in the CD measurement have an angular momentum content of 1 unit, whereas the angular momentum content of the two vortex beams used for the VCD experiment have a different angular momentum content equal to 0 and 2 units. Both i) and ii) are symmetry considerations, and they show why CD must be zero under normal incidence, and why VCD does not need to be zero. Yet, these symmetries consideration do not explain the physical mechanism that creates a non-zero VCD measurement for certain samples." On the minor note - we have changed the expression unitary vector to unit vector. We have also cited two of the latest works of Michael Berry and written the following paragraph, so that the readers know that we interchangeably write vortex beam and beam with phase singularity to mean paraxial beams which are eigenstates of the OAM operator. "Note that in general, vortex beams carrying angular momentum and beams with phase singularities are not equivalent [12,13]. But in this work, we use the terms vortex beam and beam carrying a phase singularity interchangeably because we use vortex beams with a single phase singularity of the kind exp(-i*phi).

Competing Interests: No competing interests were disclosed.

Reviewer Report 26 April 2023

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Alejandro Martínez
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Essentially, the authors use the technique unveiled in Ref. 11 to study the vectorial nature of diffraction when light impinges on objects (holes experimentally, spheres theoretically) of different sizes. Now the experiments go further to explain the surprising existence of non-zero VCD (requiring light with non-zero orbital angular momentum) on non-chiral samples when light-matter interaction is vectorial (small scatterers). The authors observe that VCD is nonzero for objects of size comparable to or smaller than the wavelength, a behavior that they attribute to the measure of vectoriality of a diffraction process. Numerical results using Mie theory also support this finding and, surprisingly, they find very similar regimes of performance (see Figs. 4a and b) in experiments and theory even though employing very different scatterers (holes vs. spheres). I think that these results are of interest to scientists doing research in light-matter interaction, in particular those working with polarized and structured light at the nanoscale. As so I would recommend indexing. Still, there are some doubts that come to my mind after reading the manuscript:

- My main concern is why VCD starts being positive (for very small scatterers) and then its sign switch to negative before tending to zero for large objects. Any explanation for this phenomenon that is consistently observed in holes and spheres?

- Does spin-orbit interaction plays any role in the whole picture? For focused beams being circularly polarized, interaction with small objects should be affected by this interaction.

- The Au layer is quite thick (200 nm). Would the results be the same using thinner Au layers (as conventionally done in plasmonics)? Does the thickness play any role?

- This sentence may be misleading: “On the one hand, the scalar regime is suitable to describe diffraction processes where the dimensions of both the beams and matter are much greater than the wavelength” since one usually relates diffraction with light illuminating objects with sizes of the order of (or smaller than) the wavelength.

Is the work clearly and accurately presented and does it cite the current literature?
Yes

Is the study design appropriate and does the work have academic merit?
Yes

Are sufficient details of methods and analysis provided to allow replication by others?
Yes

If applicable, is the statistical analysis and its interpretation appropriate?

Yes

Are all the source data underlying the results available to ensure full reproducibility?
Yes

Are the conclusions drawn adequately supported by the results?
Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Nanophotonics

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard.

Author Response 29 Apr 2024

Xavier Zambrana-Puyalto

We thank Alejandro Martínez for reviewing our article. His comments have definitely helped us to improve our work. Next, we address his points, one by one. To see a more complete response with figures, equations and references, see this PDF file attached here.

- Intuitively, we think that this effect is a consequence of the differences in beam waist of the two beams used to perform the VCD measurement. If we compare the field structure at the focus of the two different beams used for the VCD measurement, a beam with a total AM equal to 0 and another one with AM equal to 2, we observe that the field at the focus has a smaller beam waist (this can be observed in Figure 5 of the manuscript). Now, for Mie particles, we know that we usually couple more light into the resonator when the incident beam is tangential to the surface. As a result, the beam with AM=0 in-couples more light for particles with diameters below 600 nm, and the beam with AM=2 in-couples more light with diameters between 900 and 1600 nm. The z polarization component of the incident beams with l=-1 is particularly revealing for this consideration. This is shown in the figure of the pdf, where we show the z component of the two incident beams and we compare them with a circumference of diameter = 500 nm (top row) and diameter = 1200 nm (bottom row). For D = 500 nm, we observe that the beam with helicity=-1 illuminates the particle more tangentially (or via surface-scattering) than the beam with helicity=+1, whereas for D = 1200 nm is the other way around.

- We can describe light matter-interactions with different frameworks. For instance, one can describe the interaction between non-paraxial beams of light and nano-structures using the spin and orbital angular momentum. This description has been used by many authors in the field. Alternatively, one can also choose to describe the problem in terms of the total angular momentum and the handedness (or helicity) of light beams. This is what we have decided to do here, as it is a description that we are more familiar with and that we believe that it is better suited for these sort of problems. In the revised manuscript, we have included the following sentence that

reminds the readers that the problem could have been addressed within the spin-orbit interactions framework: "Notice that our theoretical modelling is based on Mie Theory and a beam description based on the total angular momentum and handedness, yet some other authors have addressed this problem in the framework of spin-orbit interactions [25-27]."

- The 200 nm layer of gold is used to make sure there is no direct transmission of the beam through the gold layer. As long as the thickness of the Au layer is such that there is no direct transmission of the laser beam, we do not foresee any big changes in the VCR(d) trend-line. However, a drastic reduction of the thickness of the layer of gold could turn the Au-glass system into a multilayer system and the VCR(d) trend-line would be affected as a result. Even if the effect of the thickness is not in the scope of this work, we have added a sentence in the main article to mention the effect of the thickness: "Note that the thickness of the Au layer has been chosen to greatly reduce the direct transmission of the beams through the sample. It is not expected that slight changes in the thickness of the gold layer will modify the results of this paper."
- We thank the referee for pointing this out. We agree that the sentence was confusing, and therefore we have changed it. Now, it reads as:
"On the one hand, the scalar regime is suitable to describe diffraction processes where *either* the beam or the matter size are much greater than the wavelength.

Competing Interests: No competing interests were disclosed.