Dichroism in a plasmonic lattice based on triskelion nanoelements

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Abstract: Dichroism is the different optical response obtained for left or right circular polarised light and characteristically occurs in chiral systems. This work is focused on enhancing the dichroism observed in the plasmonic response of Au planar nanoelements (triskelion) showing helicity. Numerical simulations have been performed to study the electromagnetic response of these systems. First, a free-standing triskelion has been studied to obtain its natural response. Then, a triskelion on a silica substrate and two stacked free-standing triskelia have been considered so as to observe the response of 3-dimensional true chiral structures. Finally, a hexagonal array of the triskelion stack has been tested to analyse the hybridisation of localised and collective modes.

I. INTRODUCTION

Plasmonics, which can be defined as the study of the response of metals to electromagnetic waves in the optical range, has recently been established as a promising field in nanoscience [1]. The interaction with the conduction electrons can configure surface and volume oscillation modes with characteristic resonance frequencies, named plasmons. Surface plasmon phenomena usually occur through the interaction of metal nanostructures with electromagnetic radiation at the interface with the surrounding dielectric medium.

The distribution of charge density associated with surface plasmons may cause the appearance of a distribution of electric field gathered near the interface that, in appropriate shape structures, can be considerably intense.

Noble metals are characterized by its interesting optical features, which can be described with a complex dielectric constant that depends on the wavelength and that usually has a negative real part. It can be seen [1] that this properties fulfill the conditions to produce localised surface plasmons, which are distinguished by an exponential decay of the associated electromagnetic field in the normal direction to the interface.

Chirality is the property described by the absence of any mirror-symmetry planes in a structure. It is key in a wide range of fields and has been determinant in science history [2]. Indeed, chirality is closely related to the optical response of some systems, as it has have been empirically found that there exists a difference in the dielectric constant for right circularly polarised light (RCP) and left circularly polarised light (LCP), for both its real and imaginary parts. A difference in the imaginary part involves a change of phase, and, consequently, light polarisation, while distinct real parts entail a difference in light absorption. Precisely, dichroism is the phenomenon that occurs when there is a difference in light absorption depending on the type of circular polarisation [2, 3]. Dichroism represents an irrefutable proof of the fundamental differences that exist upon chiral systems.

In this work, dichroism DC over any measurable magnitude will be calculated following the formula:

$$DC = \frac{I_{LCP} - I_{RCP}}{I_{LCP} + I_{RCP}} \tag{1}$$

where I_{LCP} represents any measurable magnitude for LCP light and I_{RCP} for RCP light.

In planar structures, the concept of chirality is an interesting aspect to consider. Intuitively, planar chirality can be defined as the property of any 2-dimensional object of not being able to overlap its chiral image. Nevertheless, a simple turn of the plane of the object leads to the opposite one, which implies that a planar object cannot be truly chiral. In this sense, it was found that the optical response is connected with the reciprocity concept [2]. If a reciprocal system interacts with an incident electromagnetic field, resulting in an output wave, the reverse interaction with this output electromagnetic field would result in the original signal as an output. Nevertheless, in a 2-dimensional helicoidal system the direction of rotation changes from one side to the other, so it is essentially impossible to recover the initial signal. Consequently, a 2-dimensional system can not have a chiral response, that is, dichroism. Contrarily, in helicoidal 3-dimensional objects, it can be observed an intense chiral behaviour.

The aim of this work focuses on the optimisation of the conditions giving rise to dichroism in the electromagnetic response caused by the excitation of localised surface plasmons in Au triskelion nanoelements. The triskelion is a planar structure characterised by an helicoidal symmetry which makes it chiral when reciprocity is fulfilled [4].

II. SIMULATIONS AND RESULTS

Throughout this work the electromagnetic response of several nanostructures is analysed by means of the Finite-Difference Time-Domain (FDTD) method based on Lumerical software [5]. This method is optimised for modeling nanophotonic devices and operates with

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a space-time discretisation and the direct resolution throughout this mesh of Maxwell's electromagnetic equations using finite-difference numerical integration. Additional details can be found in the appendix.

The dielectric constants for Au and silica (SiO_2) have been obtained from Johnson and Christy [6], and Palik [7], respectively

A. A free-standing triskelion

The initial step consists in the study of a free-standing triskelion in order to explore the primary response of this nanoelement.

The triskelion nanoelement is formed by three branches rotated 120° and joined each other on one side. The branches consist of an Au prism of 450 nm in length and 30 nm in both width and thickness with a 120° fold at the midpoint. Fig. 1(a) shows a top view of the triskelion.

The choice of this structure is based on its geometrical properties. The absence of mirror symmetry is a necessary condition for chirality and it is properly fulfilled. Indeed, the triskelion structure has 3-fold helicoidal symmetry, which is not compatible with the even number of electric poles that are typically formed in a multipolar excitation. Consequently, the existence of collective excitation modes could be promoted when triskelia are arranged in an ordered array.

It has been simulated the response of a free-standing triskelion being exposed to a source of RCP and LCP light, both in a spectral range between 500 nm and 1800 nm. The response has been measured through the absorption A and the scattering S cross-sections of light. In addition, the extinction cross-section E has been computed with the simple formula $E = S + A$. Light extinction has to be emphasised due to its easiness to be directly measured through light transmission.

S and A under LCP and RCP light highlight the existence of a dichroic response, which can be appreciated in the two main peaks of the two spectra, as shown in Fig. 1(c),(d). Nevertheless, dichroism vanishes in terms of E (see Fig. 1(b)), as expected for a planar structure.

The observed response in S and A correspond to, indeed, virtual dichroisms that disappear when it is considered the total optical response, that is, E. Light interacts with the structure through two mechanisms: absorption and scattering, the latter being understood as a virtual absorption followed by an instantaneous emission. It is reasonable to justify that when the triskelion is exposed to the two light polarisations, both different mechanisms occur with a significant distinct probability and, consequently, there is a dichroic response. Nevertheless, the triskelion is eminently a 2-dimensional structure, as its thickness is far more shorter that the incident light wavelength. Therefore, despite its notorious helicity, the lack of reciprocity implies the impossibility to have a true dichroic response [2]. Because of this, the dichroism observed in S and in A are complementary, cancelling each other and resulting in a non-dichroic E.

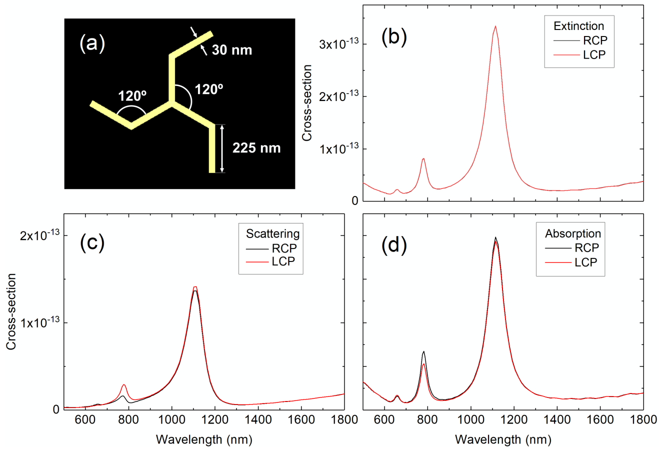

FIG. 1: Top view of the free-standing triskelion (a). Diagrams (b), (c) and (d) show, respectively, the extinction, the scattering and the absorption as a function of the wavelength for both RCP and LCP light impinging normal to the structure.

B. A triskelion on a silica substrate

To obtain a dichroic response, and accordingly to the conclusions from the free-standing triskelion, a 3-dimensional structure is needed.

A single triskelion can be converted to a 3-dimensional element by differentiating its top and bottom sides, which makes it reciprocal. This can be achieved with the introduction of a 200 nm silica layer underneath the nanoelement, as can be seen in Fig 2(a).

A simulation of an Au triskelion structure over a SiO_2 layer being exposed to RCP and LCP light has been performed. The optical response has been computed as in the previous case, and E is shown in Fig. 2(c).

If the resultant extinctions for both RCP and LCP light are compared, a small dichroic response around 800 nm and 1300 nm is found. The comparison between the current magnitude of E and that obtained for a free-standing triskelion conducts to notice that the silica layer represents the main, and major, contribution to E (through the absorption cross-section), which is reasonable taking into account the insignificant surface that the triskelion represents over the whole substrate.

In order to determine the dichroic response properly, another simulation only with the silica layer has been performed and the triskelion effective E has been calculated by subtracting the layer E from the total E that was previously computed.

As expected and shown in Fig. 2(b), dichroism is observed in the triskelion effective E, with a significant greater response for the LCP light, whose polarisation direction is opposed to the triskelion helicity. This configuration facilitates a greater S and A, which concurs with the result. However, the structure does not admit the possibility to increase this dichroic response as there is no geometric parameters to change. For this reason, it has been proceeded in a different direction in search of a configuration showing an optimal dichroic response.

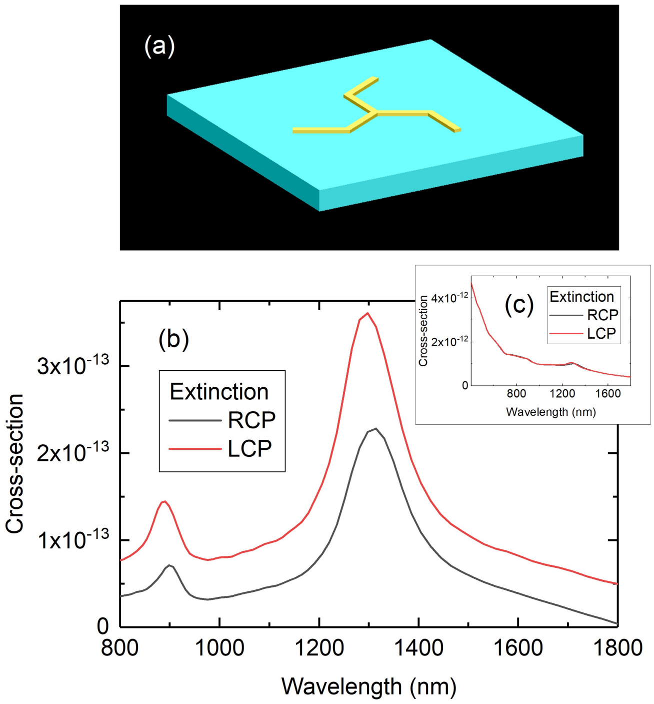

FIG. 2: Triskelion structure over a 200 nm silica (SiO_2) layer (a). Diagrams (b) and (c) show the triskelion effective E and total E, respectively, as a function of the wavelength for both RCP and LCP light impinging normal to the structure.

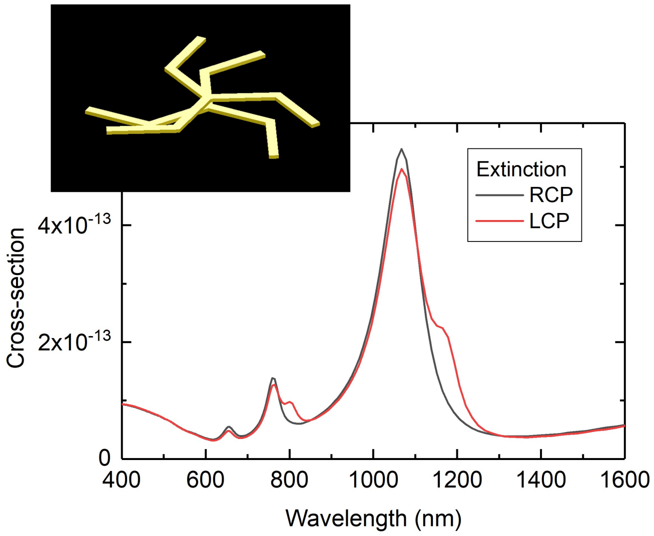

FIG. 3: Two stacked triskelia (inset). The diagram shows the triskelion total extinction as a function of the wavelength for both RCP and LCP light impinging normal to the structure.

C. Two stacked free-standing triskelia

In order to increase the chirality of the structure, a properly 3-dimensional structure has been designed. It has been proceeded with a simulation of two stacked free-standing triskelia with a separation distance of 50 nm and the above one rotated 30° counterclockwise, as shown in the inset in Fig. 3. This configuration enables to vary the distance and the rotation angle so as to modify the dichroic response at will. These parameters have been thoroughly studied in [4] and the chosen ones are adequate for a significative dichroic response.

A simulation with the free-standing Au triskelion stack exposed to RCP and LCP light has been carried out. The response has been computed as previously, that is, through measuring S and A, and the resultant E for both light polarisations is shown in Fig. 3.

The occurrence of two extra excitation modes for LCP light in the triskelion stack is observed, in comparison with the excitation spectra of the single free-standing triskelion. Apart from these modes, there is a quite general parallelism between both E spectra. It should be paid special attention to the excitation mode around 1200 nm, as it is the most intense one and will be explored in detail later.

It can be assumed that the additional E peaks for LCP light are a consequence of new excitation modes that appear due to the interaction between the two triskelia, and because of this, they are eminently 3-dimensional excitation modes. Besides, this excitation modes are apparently an intense source of dichroism.

As previously argued, the use of a 3-dimensional structure supposes an increase in the structural chirality and, consequently, enables obtaining a dichroic response. In this sense, it is reasonable to assure that those excitation modes which are truly 3-dimensional rather than planar, that is to say those that will appreciate the helicity of the structure and its chirality, will be the ones that result in a greater dichroic response, as the observed results point out.

So as to explore these new excitation modes and corroborate that they represent truly 3-dimensional excitations, it has been computed the distribution of the modulus of the electric field nearby the two stacked triskelia for the most intense additional excitation peak, around 1178 nm. To obtain a representative outlook, a simulation has been performed in which the electric field distribution has been computed in three distinct planes: one 5 nm over the top triskelion, one in the middle between the two triskelia and one 5 nm over the bottom triskelion. In Fig. 4, the distributions of the modulus of the normalised electric field are shown.

The LCP light induces an intense excitation mode in the triskelion stack, as the electric field is significantly much more intense for this light polarisation. In addition, and indeed more relevantly, it can be observed that the LCP light also induces an electric field that is notoriously significant in the middle plane between the two triskelia, while contrarily the electric field in this plane associated with the RCP light is much smaller. This corroborates the thesis that the mode under LCP light is truly 3-dimensional, as its electric field distribution shows, while the RCP light excitation is eminently planar, as it is due to an almost independent excitation of the two triskelia.

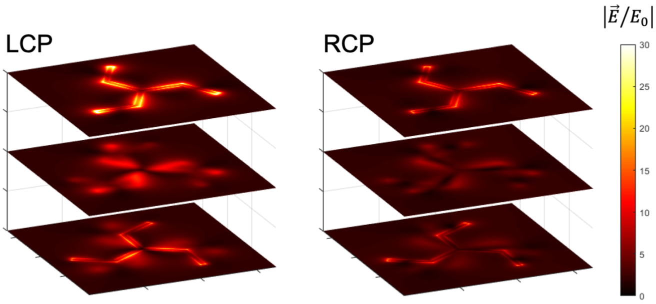

FIG. 4: Distribution of the modulus of the electric field normalised to the modulus of the incident field for 1178 nm RCP (right column) and LCP (left column) light impinging normal to the structure, measured over three horizontal planes: one 5 nm over the top triskelion (top row), one in the middle (mid row) and one 5 nm over the bottom triskelion (bottom row).

With this result it can be concluded that it has been properly designed a 3-dimensional structure that around 1178 nm exhibits excitations that differ significantly under the two circular polarisations. For RCP, it results in a planar excitation mode, which reproduces the extinction of a single free-standing triskelion, and for LCP, it results in a new intense and quite well-defined 3-dimensional excitation mode. As a consequence, it represents an important source of dichroism.

D. Hexagonal array of the triskelion stack

At this point, the triskelion stack is introduced into a lattice. The motivation of this attempt does not only come from the fact that the order of magnitude of the total response is expected to be significantly greater, as there will be a whole mesh interacting with the incident light instead of a single nanostructure, but also because it has to be considered the collective excitation modes of the lattice as a whole. In this sense, each lattice system has its own characteristic excitation modes, which could be quite useful due to its narrow spectral width and large decaying time [8].

A hexagonal lattice with a mono-element basis based on the triskelion stack has been chosen. A lattice top view is shown in Fig. 5(a). For this lattice, the condition for the collective excitation mode (analogous to the diffraction condition) for a normal incident electromagnetic wave is $\lambda = p\cos(30°)$, where λ is the incident light wavelength and p is the lattice pitch (see Fig. 5(a)). Consequently, this design allows the modification of the spectral position of the collective excitation mode by choosing an appropriate value of p.

The dichroism of the lattice has been explored by hybridising the dichroic modes previously obtained in the triskelion stack with the lattice collective ones. It is worth noting that the lattice is a 2-dimensional system, so it can be affirmed that it is not an additional source of dichroism and, consequently, the resultant response will be only an enhancement of the original dichroism.

Simulations of the optical response of a hexagonal array of Au triskelion stacks under RCP and LCP light have been carried out. Two hexagonal lattices have been set: one with $p = 1235$ nm so as to achieve a complete hybridisation with the most intense dichroic peak, and another one with $p = 1300$ nm to achieve a certain hybridisation with the same peak, but with the collective peak fairly red-shifted. Finally, the response has been measured through the total light reflection R and transmission T. Furthermore, the absorption has been calculated as $Absorption = 1 - R - T$. Results are shown in Fig. 5(b),(c).

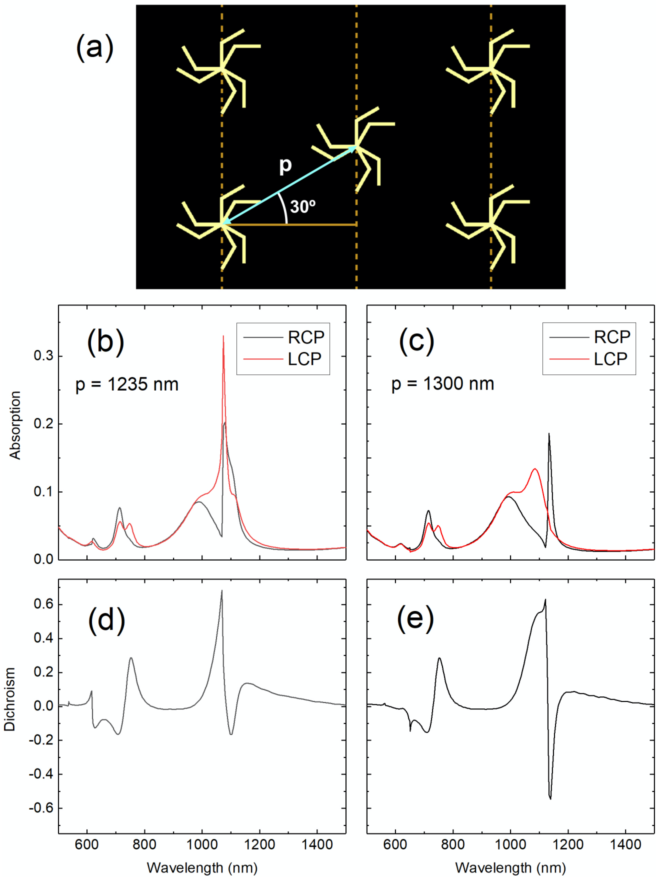

FIG. 5: Hexagonal lattice of triskelion stacks (a), where p is shown and equivalent lines of points of the Bravais lattice are represented as vertical dashed lines. Diagrams (b) and (c) show the absorption for both RCP and LCP light impinging normal to the structure for $p = 1235$ nm and $p = 1300$ nm, respectively. The corresponding dichroism computed through Eq. (1) is shown in diagrams (d) and (e).

The total absorption for $p = 1235$ nm shows that the hybridisation of the collective peak (1074 nm) with one of the localised modes of the stack (that showing 3-dimensional features) significantly increases the signal intensity and keeps the local dichroism with a considerable enhancement (see Fig. 5(b)). In addition, it can be noticed that this hybridisation results in a clear Fano resonance [9] for the RCP light, which usually occurs whenever a narrow spectral mode, such as the lattice mode, is hybridised with a broader one, such as that of the triskelion stack. Through the absorption for $p = 1300$ nm for both polarisations, two clearly different modes can be distinguished: the LCP light corresponds to a moderately more intense localised mode, as it would have been expected, and the RCP light, which goes on the stack rotation direction, to a lattice collective excitation around 1134 nm. The fact that the localised excitation inhibits the collective one induces a considerable amount of dichroism.

The absorption dichroism has been also calculated following Eq. (1) and is shown in Fig. 5(d),(e). For both pitch values it can be observed strong dichroism around the expected wavelength corresponding to the collective excitation mode, which confirms the validity of the model and corroborates the existence of a significant dichroic response from the designed lattice.

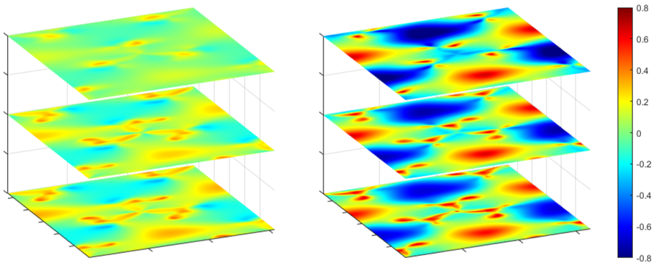

FIG. 6: Local dichroism of the modulus of the electric field computed by Eq. (1) for both $p = 1235$ nm (left column) and $p = 1300$ nm (right column) measured for 1074 nm and 1134 nm, respectively, and calculated over three horizontal planes: one 5 nm over the top triskelion (top row), one in the middle (mid row) and one 5 nm over the bottom triskelion (bottom row).

Moreover, the local dichroism of the electric field has been calculated, and the results are shown in Fig. 6. To obtain these maps of the dichroism, simulations for both pitch values have been carried out in which, as previously performed for the triskelion stack, the distribution of the modulus of the electric field has been measured in three representative horizontal planes for the wavelengths corresponding to the collective peak in each case. With the results for each structure from both RCP and LCP light, which consist in a data array that comes from the mesh that the Lumerical software implements so as to execute the simulation, the distribution of the dichroism of the electric field modulus has been calculated computing Eq. (1) locally at each cell of the resulting array.

For $p = 1235$ nm it can be observed appreciable dichroism only around each triskelia stack, from which it can be confirmed that dichroism comes mostly from a localised mode. On the contrary, the two modes observed for $p = 1300$ nm around 1134 nm are reproduced in the local dichroism representation, as it can be observed dichroism corresponding to a localised mode associated with each triskelion stack, that comes from LCP light, and dichroism distributed throughout the lattice, which corresponds to a collective mode from RCP light.

III. CONCLUSIONS

The indispensability of reciprocity in obtaining a dichroic response has been studied in a free-standing triskelion. For the stacked structure, dichroism has been successfully induced as the electromagnetic response is 3-dimensional for the polarisation that goes oppositely to the triskelion helicity. When the stack has been introduced in a hexagonal lattice, the local and collective modes have been satisfactorily hybridised yielding a significant amount of dichroism, as the collective excitation preferably appears for the circular polarisation that induces a less intense 3-dimensional excitation. Consequently, two distinguishable modes can be characterised: an eminently planar one that significantly interacts with the lattice, and a 3-dimensional localised excitation.

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APPENDIX

Simulations have been performed using the aforementioned FDTD method from the commercial software-developer Lumerical.

Lumerical integrates a graphic user interface which provides several geometric tools that enable to adjust the different parameters from the structure that is to be simulated. Two electromagnetic plane waves with a $\pi/2$ phase difference have been combined so as to obtain the circularly polarised light sources. There are also several monitors available that enable the measurement of different magnitudes, such as light intensity or the electric field modulus, which automatically normalise the results to the incident amplitude. Finally, the mesh can be configured so as to fulfill the required boundary conditions and to adjust its fineness wherever a higher resolution is desirable.

In Fig. 7 a diagram of the stacked triskelion lattice is shown within the Lumerical user interface.

Once the system is properly designed, the simulation is run with a time limit and an auto shutoff level indicator that estimates the fraction of remaining energy from the initial one and interrupts the execution in case of divergence. Indeed, it is this latter parameter which is used to indicate that the simulation has properly converged, usually around 10^{-6}.

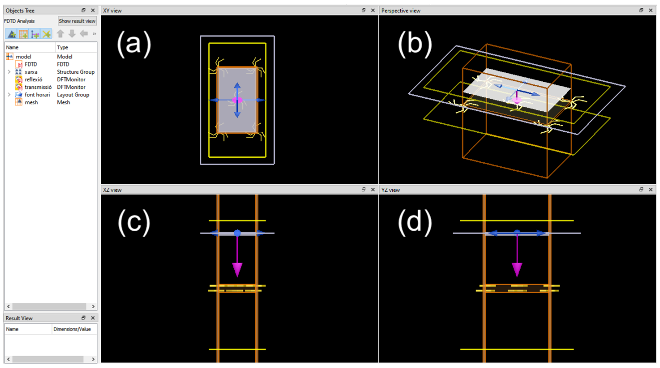

FIG. 7: Lumerical user interface, where views along the planes XY (a), XZ (c) and YZ (d) and a perspective view (b) of the designed structure are shown.

A significant amount of computing power is required to complete the proposed simulations, so the High-Performance Computing (HPC) Knowledge Base services from Consorci de Serveis Universitaris de Catalunya (CSUC) have been used, with which the simulations achieved the convergence after up to 24 hours of computation.