High Circular Dichroism through Planar Plasmonic Metasurfaces and Enhancement through Surface Plasmon Resonances

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Abstract*— Research on metasurfaces featuring high circular dichroism has been very active in the last decade, because of possible applications in various fields, such as spintronics, polarization optics and stereochemistry. Despite literature displaying various proposed geometries and methods, today it is hard to define an alternative choice combining high Circular Dichroism (CD), easy realization and robustness of performance. In this work, we show the design approach and first experimental demonstration of a planar plasmonic metamaterial based on a comma-shaped geometry, which can be used to obtain high CD. Our metasurface combines intrinsic chirality and enhancement effects due to high field localization. In particular, the relative correspondence between numerical analyses and experiments shows the possibility to tune the effects of Surface Plasmon Resonances to a certain extent. This is equivalent to the possibility of engineering stable peak values on specific wavelength ranges, even in the presence of geometrical non-idealities due to fabrication. Samples were built through standard top-down fabrication procedures, helped by the symmetry of the extrusion. The ease in implementation, combined with reliable performances, make our proposed metamaterial eligible for adoption for further research in high precision spectroscopy and optical manipulation at industrial scale.

Keywords-Circular Dichroism; Metamaterials; Plasmonics; Surface Plasmons; Microfabrication.

I. INTRODUCTION

Chirality refers to the geometric property of a structure lacking mirror symmetry planes. It exists in many forms in nature, ranging from molecules to proteins and crystals. In contrast, a structure is achiral if it is indistinguishable or superimposable on its mirror image. Due to the interest in chiral molecules, optical dichroism properties of natural and artificial materials have become a hot research topic. CD of a specific material is of interest also for other applications, such as spintronics, polarization optics and negative refractive index. It consists in the difference in optical behavior, read as transmission or absorption, to circularly polarized optical waves. Various conventions can be used to quantify CD; between all those, we prefer the approach that values more the relative relations between the two polarizations, and we put in a second order of importance the amount of transmission (this increases the alternatives):

$$CD=(T_R-T_L)/(T_R-T_L); \qquad (1)$$

where T_L and T_R are left and right polarization transmission spectra, respectively.

Literature on high CD metasurfaces is rich, and various alternatives have been developed and experimentally demonstrated [1]. Research can be categorized by the working principles (extrinsic dichroism, intrinsic dichroism, or enhancements by field localization), but, over time, the main categorization discriminant has been the fabrication method [2], because of the need to single out the most reproducible and the best performing geometries. To the great variety of geometries that have been demonstrated corresponds a big variety of fabrication techniques and approaches, both top-down and bottom-up. In general, in quantitative terms of circular dichroism factor, the top-down approach is usually more successful. Notable three dimensional (3D) nanophotonic structures are the nano-helices [3] built with focused-ion-beam-induced-deposition technique or laser writing. Stacked planar (e.g., [2]) geometries are also present in literature, and such approach also allows to accomplish strong chiroptical responses; all of them can be built by combining lithography with alignment in a layer-by-layer manner. Other than 3D and stacked 2D structures, a few simple planar structures have been successful plasmonic gammadions, nanoslit pairs, split rings, and a few others [2] can be obtained with established and standardized fabrication techniques, although with a small CD factor due to the fact that thickness is significantly lower wavelength. An interesting approach is the structure obtained by a vertical extrusion of a 2D shape, which consists in the thick deposition of a 2D patterned geometry. With this concept, it has been possible to increase the intrinsic dichroism and obtain a "giant" CD in [4]. Unfortunately, the related bandwidth is very thin and it is possible to verify that such geometry has very feeble stability, as CD peaks may vary in amplitude and central wavelength. Overall, between all the options, it is hard to find one combining high CD, reliable performances, reproducibility and accessible fabrication. In [5], the authors present a structure featuring high CD values, obtained by combining a high intrinsic dichroism due to vertical extrusion with enhancements effects due to high field localization caused by Surface Plasmon Resonances (SPRs).

In this work, we extend the work in [5] and present the related design approach and the first experimental results. We have built "comma"-shaped geometries based on noble metals (Ag and Au), after an accurate design focused on the stability of performance. The fabrication was operated through a top-down approach by using a single lithographic step to imprint the geometric shapes; the thickness of the deposited material was significant (equivalent to 1-3 tenth of the wavelength). In line-transmission measurements helped to study the performances. We also show that, to a certain extent, it is possible to control the effects of SPRs, since there is a relative convergence between numerical results and experiments.

In Section II, the available experimental works and tools are presented. Those represent the framework of the design process, which is described in Section III. In Section IV, we show and compare the numerical and experimental results and provide related discussions. In the final section, the conclusions are drawn.

II. EXPERIMENTAL METHODS AND TOOLS

Before dealing with the design, we first focus on the available experimental tools. Our goal was to define structures that can be built with standardized techniques and accessible tools. We thus decided to operate with a single step of 2D lithography (more specifically, we used Electron Beam Lithography) and lift-off process to pattern the evaporated metals. Depending on the available instruments, we decided the thicknesses of Ag and Au to be around 140 nm and 80 nm, respectively. As said previously, we fabricated two classes of metamaterials; the first was made of Ag, while the second class was made of Au, which was selected for its chemical stability and for its potential use as substrate for biological experiments.

Since the goal was to simplify the use of our metasurface, we decided to work with transmission

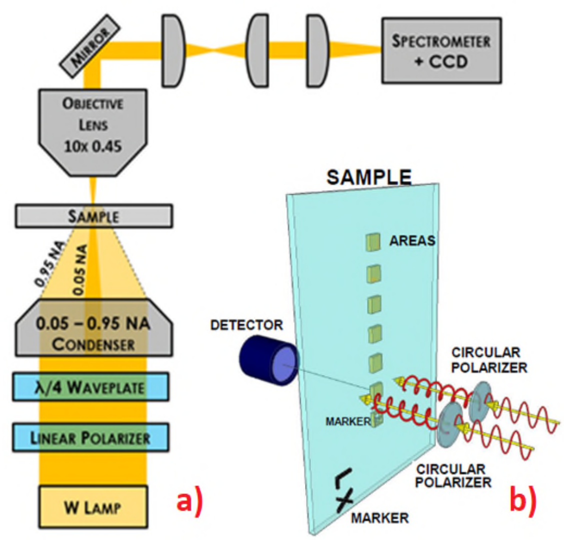

Figure 1. a) Shows the characterization setup used for the transmission analysis. The beam could reach areas down to 20 μm^2. b) Layout of the samples, which contain various areas.

measurements, which allow to use setups that are easier to integrate. That said, we still wanted to show the performances of this structure with precision, so we used the setup used in Figure 1a-b. Figure 2 shows a Scanning

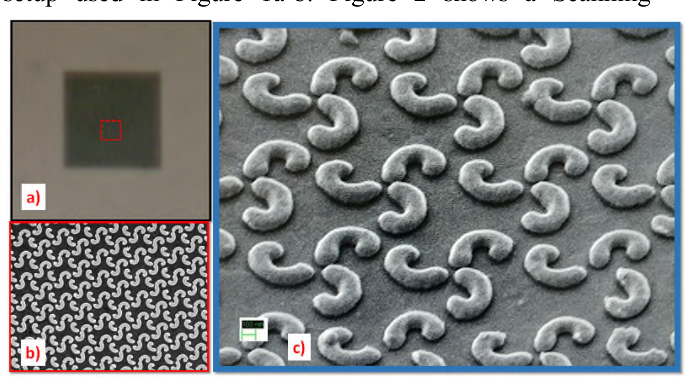

Figure 2. a) Image of an area, as seen under the microscope. b) SEM image representing the tip view. C) SEM image showing the oblique view of nano-commas; in particular, the curved lateral faces are visible.

Electron Microscopy (SEM) image of the Au sample whose experimental results are shown in this work.

III. Design Approach for Nanocommas

Having defined the fabrication and characterization requirements and limits, we then proceeded to the design of the samples. We first searched for a shape lacking sharp edges and showing a degree of intrinsic CD; the avoidance of edges helps to reduce the variability of performances and eases the fabrication requirements. Between the plethora of possible geometries, we selected those which could be easily replicated with standard Electron Beam Lithography (EBL) processes, and could be distributed with high density on a surface. After some tests with the EBL, we heuristically chose a comma shaped geometry.

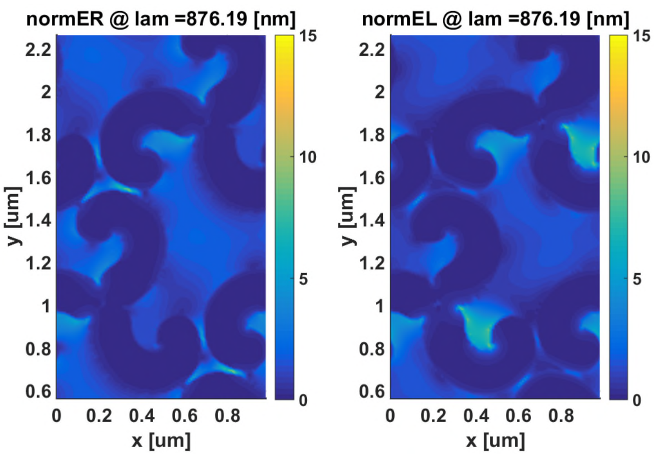

Figure 3. Example of field localization of the two types of polarization. Those concentrate on different sections of the nanocommas, depending on the circular polarization of the light (top is Right, bottom is Left).

The metamaterial in sample Ag was created after setting the “commas” in “triplets”, and those were then further inserted in triangular super-grids. The numerical methods and the specific operations used for the design are shown in [5], and the resulting grid cell is shown in Figure 3, along with an example of the field localization.

The Au sample was designed with a further evolution of the previous approach, as shown in [5]. This time, the “comma” shape was represented with the geometrical parameters of Archimedean spirals, and those parameters were weighted with their estimated variability after fabrication. Various numerical analyses were performed in order to find equilibrium situations where CD peaks were relatively constant.

IV. Results And Discussions

Some results from the Ag sample are shown in Figure 4. As can be seen, even though the resulting CD is relatively high, the amplitude varies significantly when different surfaces are tested (the regions shown in the image represent the tested positions). This means that there is a strong variability of the performance, even in the presence of small variations. Still, the central wavelength of the peaks is conserved, and this satisfies one of the main objectives of the design.

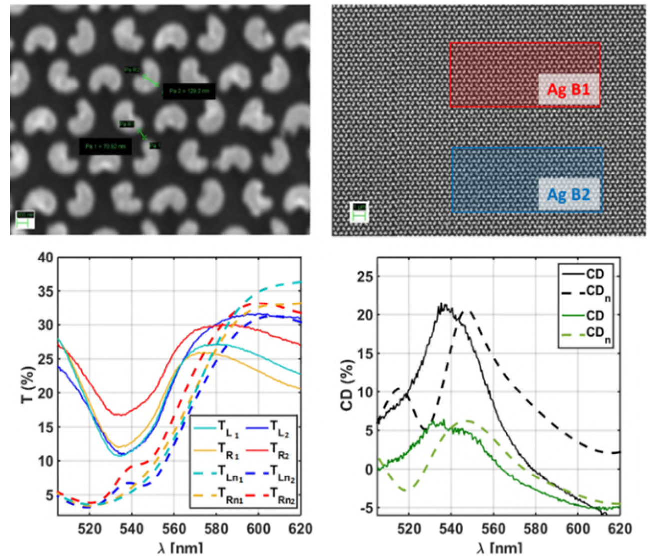

Figure 4. Top-Left: SEM image of the Ag sample’s layout Top-Right:.A symbolic representation of the characterization on two different surfaces in the same metamaterial area (which should have the same geometry). Bottom-Left: Transmission of the two polarizations according to numerical methods (dashed lines) and experiments (full lines); areas B1 and B2 are represented with indexes 1 and 2. Bottom-Right: Circular Dichroism calculated numerically (dashed lines) vs. experiments (full line); black lines correspond to area B1, while green lines correspond to area B2.

An extract from the first Au sample’s characterization is shown in Figure 5. As can be seen, the peaks are stable, since they are similar in amplitude and the peaks are around the same central wavelength. Unfortunately, the amplitudes of the CD peaks are lower than the expectations. This is probably due to the fact that this sample, as shown in Figure 1c, has round lateral profile due to a non-ideality in fabrication, while our design expected an extrusion of the same shape. Thus, the resonance at different heights is different, and this does not help to compose an overall strong CD.

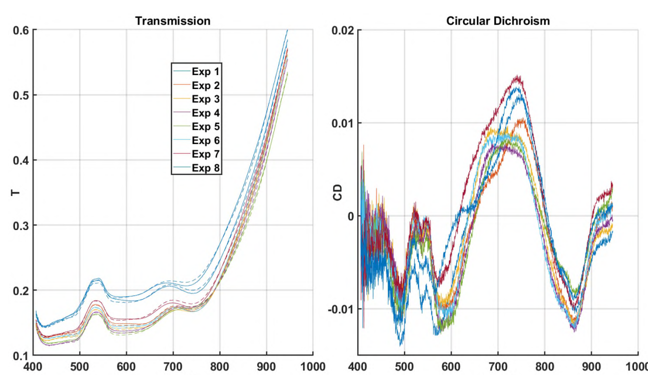

Figure 5. Left: Transmission of the two polarizations according to numerical methods and experiments. Right: Circular Dichroism calculated numerically vs. experiments

V. CONCLUSIONS

In this work, we have shown that Surface Plasmon based metamaterials may reach high levels of linear CD, if we combine intrinsic chirality with enhancements due to Surface Plasmon Resonances. Our structure can be obtained with standard planar fabrication processes and involves the vertical extrusion of a planar shape, which has soft edges.

The design process succeeded in fixing the central wavelength, although the heuristic approach of the Ag sample revealed a variable amplitude depending on the lit area, while the Au sample revealed a more stable behavior, as per design. Thus, the parametrization of the geometry and the weighted sweep can be seen as viable strategy. Unfortunately, the Au sample presented here had a relatively small CD, due to a limited thickness and a curved lateral profile, as shown in oblique SEM images.

Overall, this class of metamaterials has shown potentialities and deserves more attention and research. In particular, Au samples with higher thickness values are now under investigation. The relative correspondence between numerical analyses and experiments shows the possibility to further develop this design method, which consists mainly in finding stable peaks in Surface Plasmon Resonances. The reliability in results and the repeatability and scalability of this metasurface makes it interesting for its adoption for further research or industrial applications.

REFERENCES

[1] Z. Wang, F. Cheng, T. Winsor and Y. Liu, “Optical chiral metamaterials: a review of the fundamentals, fabrication methods and applications”, Nanotechnology, vol. 27, pp. 412001-412021, 2016.

[2] T. Tanaka and A. Ishikawa, “Towards three-dimensional optical metamaterials”, Nano Convergence, vol. 4, is. 34, 2017.

[3] A. Benedetti et al., “Precise detection of circular dichroism in a cluster of nano-helices by photoacoustic measurementsBenedetti”, Scientific reports, vol. 7 , pp. 5257, 2017.

[4] A. Y. Zhu et al., “Giant intrinsic chiro-opticalactivity in planar dielectric nanostructures,” Light. Sci. & Appl., vol. 7, pp. 17158, 2018.

[5] A. Veroli et al., “Planar chiral plasmonic 2d metamaterial: Design and fabrication”, AIP Conference Proceedings, vol. 2145, pp. 020015, 2019.