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Dielectric multilayers for broadband optical rotation enhancement

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Summary. — We design a simple dielectric multilayer capable of sustaining broadband superchiral surface waves. We show that the platform can produce large optical chirality enhancements in a wavelength range of hundreds of nanometers. We finally demonstrate that these properties result in the enhancement of the optical rotation signal well above two orders of magnitude, thus extending surface-enhanced chiral spectroscopies beyond the traditionally addressed circular dichroism signals.

1. – Introduction

Chirality is the geometrical property of three-dimensional objects that are not superimposable with their mirror image. In this context, two identical objects of opposite chirality are called enantiomers. Chirality plays a pivotal role in biochemistry and is found everywhere in biomolecules, with DNA being the most notable example. Therefore, the separation and analysis of chiral molecules have gained significant traction in the pharmaceutical and biochemical industry in recent times.

Chiral spectroscopies, *i.e.*, the probing of the differential interaction between chiral matter and left (L) and right (R) circularly polarized light (CPL), are among the most important means of enantiomer investigation. However, spectroscopic chiral signatures are normally feeble if compared to those of standard spectroscopy, making the investigation of small enantiomer volumes very problematic. Recently, novel methods have been

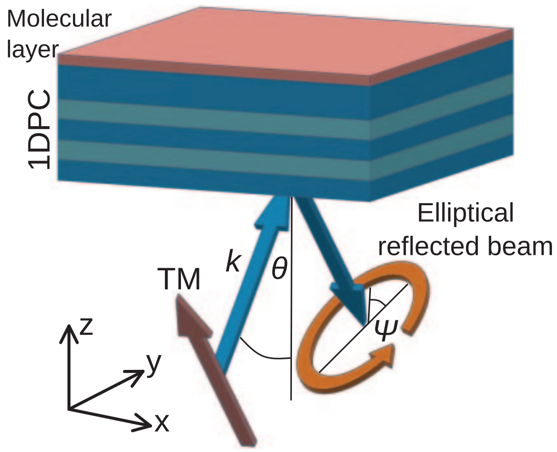

Fig. 1. – Schematics for the multilayer platform and the illumination configurations.

suggested to enhance the chiral signal by tailoring the optical chirality of the probing electromagnetic field, where the optical chirality C is defined as $C = -\frac{\varepsilon_0\omega}{2}\mathrm{Im}(\mathbf{E}^* \cdot \mathbf{B})$. This quantity behaves as a chiral object under mirror inversion, and determines the degree of asymmetry in the interaction of a chiral molecule with L and R circularly polarized light. Circularly polarized plane waves are the prime example of a chiral field configuration, where the chirality of a unitary amplitude wave is expressed as $C^{\mathrm{R,L}}_{\mathrm{CPL}} = -\frac{\varepsilon_0\omega}{2c}$. Accordingly, an electromagnetic field with C larger than the $C^{\mathrm{R,L}}_{\mathrm{CPL}}$ value is defined as being "superchiral" [1].

A large variety of plasmonic and dielectric nanostructures has been introduced and employed as viable solutions for the enhancement of the optical chiral response of biomolecules. Despite the encouraging recent results, most of the work carried out until now has been solely focused on the enhancement of the circular dichroism (CD) signal, *i.e.*, the differential absorption between left and right CPL [1-3]. On the other hand, the phenomenon of circular birefringence (CB), also known as optical rotation (ψ), involves the differential phase accumulation between left and right circularly polarized light, and results in the rotation of the plane of incidence of a linearly polarized plane wave. However, contrary to his absorption counterpart, the enhancement of this phenomenon has largely been overlooked.

2. – Results

To address this issue, we model a simple dielectric multilayer structure capable of supporting Bloch surface waves, *i.e.*, waves at the interface of a semi-infinite one-dimensional photonics crystal (1DPC) and existing in both transverse electric (TE) and transverse magnetic (TM) polarization states. We tune the thicknesses of our multilayer in such a way that the dispersion relations of the TE and TM modes are degenerate and superimposed, thus leading to the creation of a chiral surface wave (CSW), in close analogy to the traditional CPL plane waves [3-6].

A sketch of a sample design, illuminated in a total internal reflection (TIR) configuration, is shown in fig. 1. The structure consists of 5 layers of alternating low (n_{L}) and high (n_{H}) refractive index materials. We choose SiO$_2$ ($n_{\mathrm{L}} = 1.454 + 0.0001i$) and

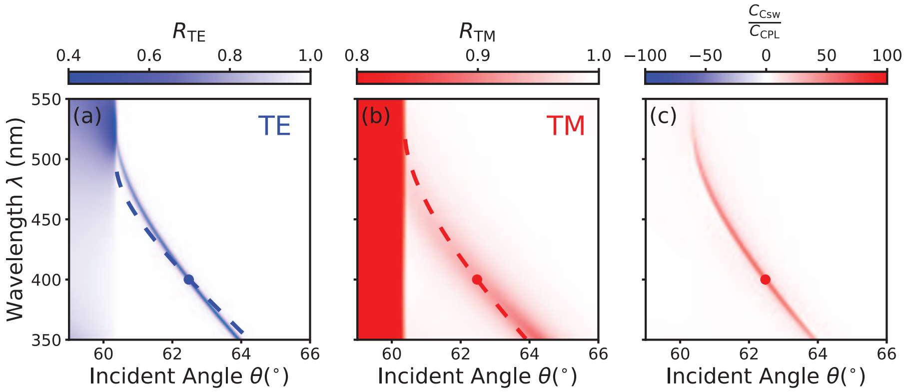

Fig. 2. – (a) TE reflectance map, (b) TM reflectance and (c) optical chirality enhancement map. The dot highlights the crossing of the dispersion relations at $\lambda \sim 400$ nm.

Ta_2O_5 ($n_H = 2.06 + 0.001i$), and tune the five layer thicknesses as $\mathbf{t} = \{t_L = 198$ nm, $t_H = 62$ nm, $t_L = 225$ nm, $t_H = 64$ nm, $t_L = 246$ nm$\}$. We then top the dielectric structure with a 5 nm thick chiral multilayer with an electronic transition at $\lambda = 400$ nm, as described in ref. [3]. Finally, we illuminate the multilayer from a BK7 glass substrate ($n_{sub} = 1.53$) and choose water ($n_{sup} = 1.33$) as the structure superstrate.

Figures 2(a) and (b) report the reflection maps as a function of the angle of incidence θ and the wavelength λ for both polarization states of the illumination. The BSW modes are clearly visible as a sharp blue band (TE) and a cloudy red halo (TM) crossing diagonally the figures from the top left to the bottom right, while total internal reflection occurs below $\theta \sim 60°$. The mode overlapping is illustrated by the dashed lines, which highlight the TM (TE) mode position in the TE (TM) map. More information is provided by fig. 2(c), which reports the (θ, λ) map of the optical chirality enhancement defined as $\frac{C_{CSW}}{C_{CPL}}$ and probed 5 nm above the multilayer surface. A strong maximum is visible at the superposition of the TE and TM modes, with optical chirality enhancements of two orders of magnitude. This clearly illustrate how the platform can sustain strong superchirality in a wavelength range of hundreds of nanometers.

Once established that the designed structure can efficiently sustain superchiral electromagnetic fields, we wish to investigate how the achieved superchirality influences the platform optical rotation signals. In the following, we shall define the optical rotation

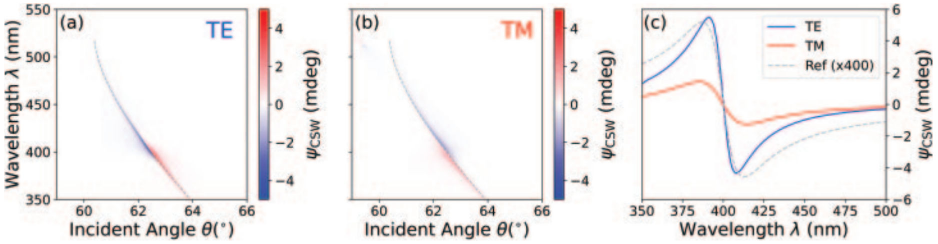

Fig. 3. – (a) TE optical rotation map, (b) TM optical rotation map and (c) optical rotation cross section along the BSW mode superposition.

as $\psi = \frac{1}{2}\arctan(S_2/S_1)$, where S_2 and S_1 are the Stokes parameters of the collected radiation [4]. We compute the optical rotation for our superchiral platform and, for comparison, also for a thin, normally illuminated 5 nm molecular layer deposited on a bare glass substrate [4]. Figures 3(a) and (b) illustrate the optical rotation (θ, λ) maps for the multilayer when illuminated by the TE and TM incident polarization states. It is immediately clear that a strong signal emerges exclusively at the superposition of the dispersion relations of the BSWs. In particular, a strong positive signal is present below $\lambda = 400$ nm, with a sign reversal for longer wavelengths, recalling the characteristic sigmoidal shape which is usually observed in rotation spectra at resonance [4]. A spectral cross section taken along the superposition of the surface modes (fig. 3(c)) indeed reveals that the multilayer platform can produce an optical rotation enhancement of $\psi_{\mathrm{CSW,TE}}/\psi_{\mathrm{CPL}} \sim 400$ for TE illumination and $\psi_{\mathrm{CSW,TM}}/\psi_{\mathrm{CPL}} \sim 100$ for TM illumination, clearly illustrating the potential of multilayer dielectric structures regarding their application for surface enhanced chiral spectroscopies. We finally notice that, while preserving the typical sigmoidal structure, the enhanced signal is slightly distorted if compared to the original one. The distortion occurs because of two different and opposite contributions to the enhancement mechanism. On one hand, the molecular absorption at $\lambda \sim 400$ nm dampens the BSW modes and leads to a smaller enhancement. Conversely, the optical chirality enhancement is larger at $\lambda \sim 400$ nm, because the BSW are perfectly superimposed at that wavelength, and slightly offset at lower and higher energy. Overall, the last contribution is more relevant, leading to a small sensitivity degradation as we move away from the BSW mode crossing at $\lambda \sim 400$ nm, where the chirality enhancement is the largest.

3. – Conclusions

In conclusion, we have shown that a simple dielectric multilayer design is capable of supporting superchiral surface waves over a broad wavelength range. The present structure allows for optical chirality and rotation enhancements above the two orders of magnitude. This opens a new perspective regarding the application of surface enhanced chiroptical spectroscopy for the analysis of low volumes of chiral molecules.

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