Origin of extrinsic chirality in metasurfaces and nanoholes fabricated by nanosphere lithography

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Abstract. Nanosphere lithography is a cost- and time-efficient tool for the fabrication of various nanostructured materials. Multiple steps of metal layer deposition at different oblique angles were shown to produce complex asymmetric and chiral shapes. Here, we investigate samples in which polystyrene nanospheres are covered by Ag or combination of Ag and Au at a single step (under 45°). In this way, we obtain metasurfaces with asymmetric shells, with a nanohole array formed due to the shadowing effect. We investigate chiro-optical properties of four samples by exciting them in the 700-1000 nm range, at angles of incidence from -45° to +45°; we report on dissymmetry in the total extinction between left and right circularly polarized excitation g_{ext}, which follows the rules of extrinsic chirality. We then resolve the transmission of Ag metasurface in terms of hyperspectral Stokes parameters, and we connect the S_3 parameter with g_{ext}. Finally, we characterize nanohole arrays obtained from the same samples when the nanospheres are removed; we further perform electromagnetic simulations to gain insight into the "egg" shaped nanohole.

1 Introduction

Modern nanophotonics relies on light-matter interaction tailored by specially designed materials at the nanoscale. Chirality in nanostructures is one of its branches, which treats the symmetry breaking at the nanoscale. Optical chirality is closely related to the dissymmetry in the interaction with the circular polarizations of opposite handedness, which finds applications in chiral sensing [1], holographic displays [2], nonlinear effects [3], to name a few.

Low-cost self-assembling nanofabrication such as nanosphere lithography (NSL) can be used to lower the cost of chiral nanophotonics. For example, oblique angle deposition of metallic layers in multiple steps led to intrinsically chiral nanocrescents [4], and nanohole arrays [5]. Moreover, chirality can be induced by properly orienting the asymmetric nanostructure with respect to the excitation/emission measurement set-up, leading to "extrinsic" chiral behavior [6,7].

We recently demonstrated rich resonant behaviour in NSL-based metasurfaces asymmetrically covered by Au [8]: polystyrene nanospheres (PNS) are periodically organized and covered by asymmetric shells, while the nanohole array (NHA) is formed on the glass substrate. In this work [9], we use similar extinction-based experimental set-up to measure extrinsic chirality in Ag- and Ag/Au-based samples. We further resolve the polarization state of the output beam under linear excitation. Finally, we investigate the NHA-only samples. Detailed electromagnetic modelling is further needed to reveal the interplay between extrinsic and intrinsic effects, and the influence of the geometric parameter margins.

2 Extrinsic chirality characterization

Here we present results for the sample based on PSN of 350 nm diameter, in 2D arrangement with periodicity of 518 nm; the PSN were covered at 45° by 55 nm Ag. We first measure total extinction when Ag-based metasurface is excited with left or right circular polarization (LCP or RCP, respectively), under angle θ.

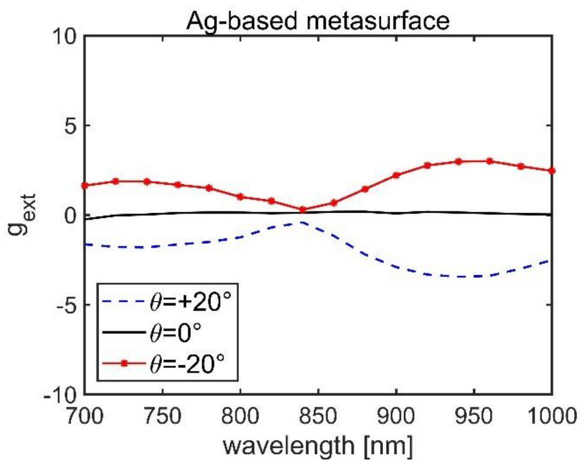

Fig. 1. Ag-based metasurface exhibits handedness-dependent total extinction at oblique incidence.

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Extinction dissymmetry factor is defined as $g_{ext}[\%]=100(Ext_{LCP}-Ext_{RCP})/(Ext_{LCP}+Ext_{RCP})$, where Ext stands for extinction. Fig. 1 plots the response at angles 0° and ±20°; as expected, chirality inverts sign with inverted θ.

We next perform full broadband Stokes parameters analysis of the beam transmitted through the metasurface when it is excited with linear polarization. In Fig. 2 we show S1, S2 and S3 as a function of θ at the wavelength of g_{ext} maximum, i.e. 940 nm. S3 indeed inverts sign for opposite angles in a wide range of angles. In particular, maximum absolute value of S3 occurs at θ=±20°, meaning that at this incidence angle the output polarization is converted from linear to the elliptical one. This is in agreement with the behaviour in extinction and absorption (not shown here).

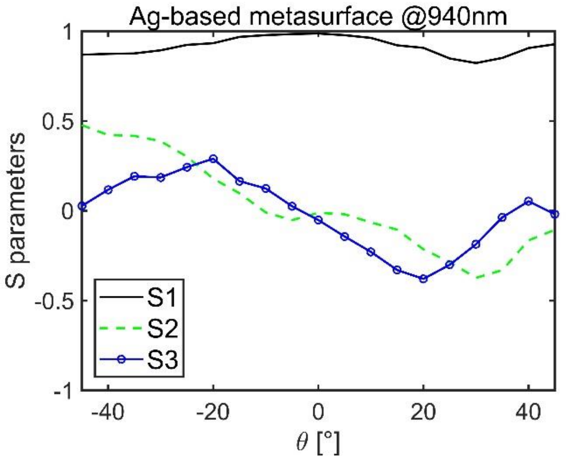

Fig. 2. Ag-based metasurface, excited with linear polarization at 940 nm, exhibits angle-dependent Stokes parameters.

We next investigate NHA-based sample where the polystyrene nanospheres are removed. The form of g_{ext} is again inverted in the near-infrared range, leading to rather low circular dichroism (CD), Fig. 3.

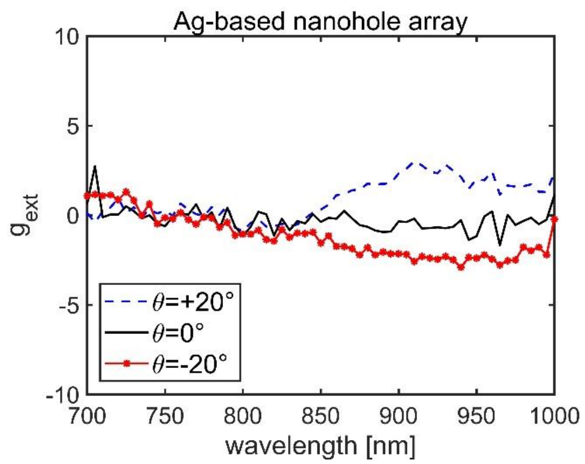

Fig. 3. Ag-based metasurface exhibits handedness-dependent total extinction at oblique incidence.

We note that similar nanostructures were proposed for optimized chiral properties in the same range [10]. However, for sensing applications, Ag should not be directly exposed to the solvent. We therefore checked the influence of Ag-Au combination on the CD behaviour: in comparison to all other geometric parameters, it results negligible. Therefore, to protect the plasmonic behaviour of Ag, we propose to cover the sample with a thin layer of Au.

3 Towards optimization

We further use a commercial 3D Maxwell equation solver to better understand the electromagnetic field confinement around the nanostructures, and the origin of the symmetry breaking in different wavelength-θ configurations. Especially interesting is that due to the shadowing effect, NHA appear in "egg"-like shapes. Such oval nanogeometries were recently proposed for BICs [11]. We believe that the numerical optimization will help in obtaining better CD factors in these simple plasmonic 2D substrates, to be further applied in sensing and emission applications.

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