The broadband enhanced chirality revealed by broken L-shape metamaterial platform

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Abstract: We experimentally demonstrate and theoretically verify a spectrally controllable, extremely large, broadband chiroptical response from three-dimensional all-dielectric broken L-shape nano-boomenrang metamaterial platforms. This innovative design holds great potential for seamless integration into on-chip photonic devices.

Main Text:

Chirality, often described as handedness is an intriguing inherent property of certain materials and it denotes an object's quality that cannot be superimposed onto its mirror image through a series of simple symmetry operations. This symmetry breaking phenomenon has garnered remarkable attention recently due to its diverse applications in polarization optics, sensing, catalysis, stereochemistry, quantum information systems, biological imaging, and spintronics [1-3]. Circular dichroism (CD), is the optical manifestation of chirality phenomenon, the differential absorption (A) ability of right-handed circularly polarized light (RCP) from that of left-handed circularly polarized light (LCP). Chirality in natural materials often exhibits weak CD values, lacks tunability, and is generally found in the deep ultraviolet (UV) part of the spectrum. Metamaterials, featuring subwavelength-scale structures, are frequently proposed as a promising solution to achieve strong and controllable chirality [1-5]. However, challenges persist in both the fabrication and chiroptical characterization of such structures. As an emerging large-scale area, bottom-up 3D nanomorphology fabrication method, *glancing angle deposition* (GLAD) is envisioned as a promising route to the experimental realization of strong and tunable chiroptical responses. Basically, this physical vapour deposition technique utilizes a programmable substrate manipulator and highly collimated particle flux that impinges upon sample substrate at extremely oblique angle [1,2].

Here, we introduce a new metamaterial design fabricated by custom-built, ultra-high vacuum GLAD technique using a two-step process. Initially, a nano-columnar array is deposited on a glass substrate from a silicon (Si) particle flux. Subsequently, the sample stage undergoes azimuthal rotation, denoted as the distortion angle (β), prior to the deposition of a second layer, which is also composed of silicon. To prevent any nonidealities in the structure design, the electronic shutter mechanism is utilized during pre-deposition substrate rotation process. The resulting simplistic metamaterial design is called as self organized, spatially coherent, three dimensional, super-lattice type, all-dielectric broken L-shape so-called *nano-boomerang* metamaterial platform. Depeding on the sign of distortion angle, the handedness of chiral response can be determined. Figure 1a shows the high resolution scanning electron microscopy (HR-SEM) images of LH and RH metamaterials. While we predict that this new metamaterial class exhibits chiroptical activity, it is known that well-oriented columns possess various other optical anisotropies (e.g. circular birefringence, linear dichroism), as well [1,2,6]. Hence, to tackle with the measurement problem of chirality for metamaterials with pronounced non-chiral properties and semi-transparent nature, we utilized generalized spectroscopic ellipsometry in Mueller matrix configuration which operates in both transmission and also reflection so that one can populate and also differentitate chirality from other optical anisotropies.

Recently, chirality response has reported in terms Kuhn's dissymmetry factor which is the ratio of CD to the sum of absorption (g_K=2CD/(A_{LCP}+A_{RCP})) [1]. This new metric makes the comparison of chirality possible for different structures independent from their fabrication and characterization techniques. Figure 1 (b) shows Kuhn's disymmetry factor spectrums of LH (red line) and RH (blue line) structures. The presence of structural mirror symmetry between LH and RH L-shaped metamaterials (see HR-SEM images in Fig. 1 (a)) is clearly projected on the experimentally measured positive and negative dissymmetry factors, respectively, in Fig. 1(b). Negligible differences between the two symmetric chiral responses depicted in Fig. 2(b) are attributed to

imperfections during the GLAD fabrication process, such as the fanning effect due to the anisotropic broadening growth of nanopillars [2]. The experimentally obtained g-factor values are very high and clearly demonstrate the strong and ultra-broadband chirality of the presented metamaterials.
As it is seen from the finite element modeling (FEM) based theoretical systematic study, we provide full map of chirality as a function of distortion angle which shows the chirality-switching response based on just by varying the distortion angle of the fabricated metamaterials. Our theoretical findings were also experimentally verified by experiments. In order to further investigate the electromagnetic modes responsible for the structurally induced chirality, we also obtained the scattering dichroism ($\Delta S = S_{RCP} - S_{LCP}$) and also is decomposed into its multipole expansion under both left- and right-handed circularly polarized illumination. Thereby, the contribution of individual underlying electromagnetic modes on chirality is also determined. Another unique property of this new metamaterial design is to control extrama point of chirality with minimum material addition as it is compared with other metamaterial design in the literature [2-5],[7-9].

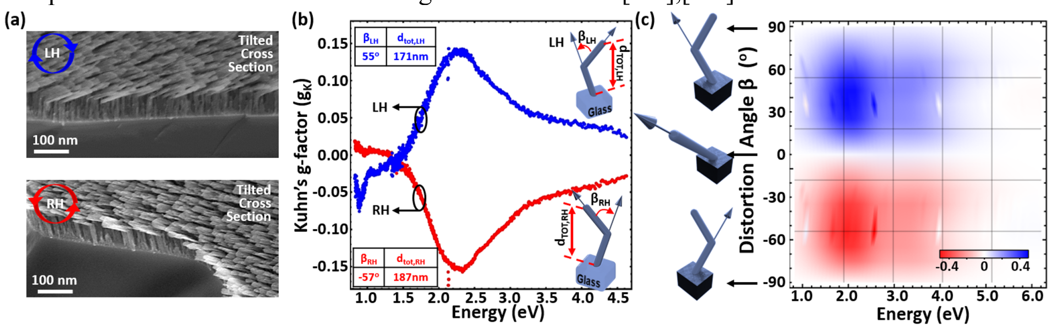

Figure 1 (a) High-resolution tilted-cross section scanning electronc microscopy images of left-handed(LH,top) right handed (RH, bottom) all-dielectric L-shape metamaterial design. (b) The combined near normal reflection and straight through transmission modes Mueller matrix spectroscopic ellipsometry based Kuhn's dissymmetry factor spectrums of RH (red color) and LH (blue color) metamterials. Inset tables show the distortion angle (β_i i:RH or LH) and total thickness of the structure ($d_{TOT,i}$ i:RH or LH) from anisotropic Bruggeman effective medium approach based generalized spectroscopic ellipsometry data analysis. (c)FEM based theoretically computed spectral evolution of Kuhn's dissymmetry factor as a function distortion angle.

To sum up, the proposed large scale-area *nano-boomerang* metamaterial design from achiral Si nanocolumnar segments exhibits among the highest and broadest experimentally measured chiral response within the wide spectral range of near-IR to UV frequencies making them advantageous compared to other relevant structures [2-5],[7-9]. With its simplistic design and scalable fabrication, it is envisioned as an ideal platform for seamless integration, particularly into on-chip photonic integrated circuit designs, next-generation sensing devices, and quantum information systems, for example.

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