

Construction of 3D Plasmonic Chiral Nanostructures on DNA Template

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The asset of DNA as nature's designer toolkit for structural technology has been explored for several decades. The excellent control over topology and superior accuracy in templated synthesis bestow DNA the most successful molecule for programmable assembly of matter on the nanoscale. Significant progress in DNA nanotechnology has been concurrent with the prompt spanning of the scope of utility and diversity of possible conjugate materials.

Recently, considerable effort has been directed towards DNA origami templated assembly of gold nanoparticles into assortments of functional nanostructures. Our work has realized the precise organization of gold spherical nanoparticles into a helical chain on a DNA origami template.¹ Optical chirality in the visible spectral range is one of the important pursuits in that it generally does not occur in natural chiral molecules. We demonstrate that circular dichroism can be generated with artificial plasmonic chiral nanostructures composed of the minimum number of spherical gold nanoparticles required for 3D chirality.² We utilize a rigid addressable DNA origami template to precisely organize four nominally identical gold nanoparticles into a three-dimensional asymmetric tetramer (Figure 1a). Due to the chiral structural symmetry and the strong plasmonic resonant coupling between the gold nanoparticles, the 3D plasmonic assemblies undergo different interactions with left and right circularly polarized light, leading to pronounced circular dichroism. We showed that gold nanorod (AuNR) 3D plasmonic chiral colloids can also be assembled by DNA origami (see Figure 1b).³

The programmability of DNA offers unprecedented spatial control over discrete constituents down to the nanoscale. Simultaneously, this capacity also provides an excellent playground to explore exotic optical functionalities of artificial nanostructures, in particular, chirality at optical frequencies. The ubiquitous relationship between the 3D configurations of plasmonic chiral objects and their chirality will be the driving force to develop a new generation of 3D plasmon rulers, which hold great potential for structural analysis, pharmacology, and biomachinery.

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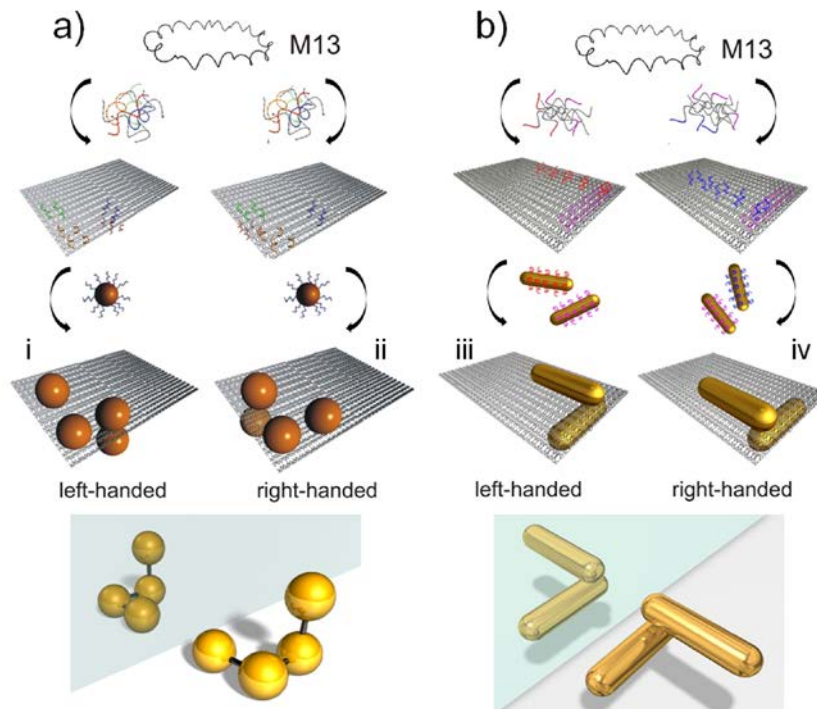


Figure 1: Schematic illustration of the experiment: A long scaffold single DNA strand (M13) hybridizes with helper and capture strands to form a rectangular DNA origami template with well-defined binding sites. a) The capture strands are extended from four binding sites, three on the top surface and one on the bottom surface of the origami template. These four binding sites are arranged in left- and right-handed configurations. Gold nanoparticles (AuNPs, 22 nm) functionalized with corresponding complementary DNA strands are assembled at the pre-designated locations on the origami template through DNA hybridization, forming left- (i) and right- handed (ii) structures, respectively. b) The capture strands are extended from the top and bottom surfaces of the origami template. Multiple binding sites are used to robustly assemble one gold nanorod (AuNR). The binding sites with different DNA sequences are illustrated using red, purple, and blue colors. AuNRs (nominally 40 nm×12 nm) functionalized with corresponding complementary DNA strands are assembled at the pre-designated locations on the origami template through DNA hybridization, forming left- (iii) and right- handed (iv) structures, respectively.