Artificial chirality evolution in micro-/nano-scale 3D plasmonic metamaterials

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Plasmonic chiral metamaterials has attracted significant attention at it provides a new route to intriguing optical properties such as negative refractive index, light polarization filters, and phase modulation. However, the fabrication regarding with limited resolution in conventional synthesis and complexity of asymmetric synthesis pose a major hindrance for further development. In this study, a new class of three dimensional chiral plasmonic nanostructures was successfully fabricated using molecular shape modifiers and crystallographic control of nanoparticle. Previously, we developed three-dimensional chiral metamaterials based on photolithography overlay and electron-beam lithography overlay for terahertz and near-infrared, respectively. We found scaling down them further to visible frequency is extremely difficult. As an alternative solution, we discovered a novel system that characteristic of molecule is transformed into distinctive gold nanoparticle shape. On the basis of this system, chirality transfer between molecular modifier and gold surface allow us to achieve numerous chiral morphologies of gold nanoparticle, named plasmonic helicoids. Particularly, enantiospecific interaction of molecule and high index plane plays pivotal role to provide asymmetric structuring process on the gold surface, forming distinct chiral morphology in single nanoparticle level. One of the representative shapes of helicoid structure showed gammadion-like structure, consisting of four highly curved arms of increasing width, in all six faces of cubic geometry. The unprecedented chiral morphology of plasmonic helicoid has remarkable optical activity (dissymmetry factor ~ 0.2 at 622 nm) even in a randomly dispersed solution, substantiated by direct visualization of macroscopic color transformation. Changes in molecular recognition and growth parameter led to different morphological evolution, and structural alterations provided a straightforward means of tailoring optical response, such as optical activity, handedness, and resonance wavelength. Also, our aqueous phase synthesis is readily scalable without losing exquisite chiral structure at nanoscale. In these aspects, our approach, chirality evolution in single nanoparticle, provides a truly new paradigm and valuable insight for chiral metamaterial fabrication. Such unique fabrication technique will provide the opportunity to achieve the significant step making metamaterials from science to technology.

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