

Abstract

Light-matter interactions at the nanoscale have been widely studied over the past few decades. In particular, the interaction of light with asymmetric nanostructures has harbored the interests of chemists, biologists and physicists alike. The world around us is largely constituted of asymmetric structures such as DNA, sugars, amino-acids, proteins, enzymes which form the backbone of every living matter. Structures which cannot be superimposed on their mirror images are termed as *chiral* structures. Naturally occurring chiral objects display unique optical properties such as Circular Dichroism (CD) and Optical Rotation, although these effects are typically very weak and occur in the UV.

In recent years, researchers have focused in designing artificial chiral substrates with large chiral response in the visible, which are orders of magnitude stronger than the naturally chiral objects. These engineered systems are suitable for a wide range of applications such as broadband circular polarizers, chiral molecule detection and negative refractive index media. The design scheme for chiro-plasmonic systems relied on the assembling plasmonic achiral nanostructures in chiral geometries or fabricating plasmonic materials of chiral geometries. In the work presented in this thesis, we present a detailed theoretical and experimental investigation of plasmonic effects in different two and three dimensional chiral systems.

One of the design schemes proposed in this work consists of vertical stacking of oppositely handed 2D chiral structures. Owing to the strong plasmon coupling between the individual nanostructures, there was a significant enhancement in the calculated CD values as opposed

to the isolated planar components. Varying the separation and the relative orientation of the layers rendered the optical response tunable in the visible. The other strategy proposed here was placing an achiral plasmonic NP in chiral hotspot of the chiral plasmonic structures. The results from the numerical simulations suggest the interaction between a chiral and achiral NP at close proximity could be a way for enhancing the chiral response in the visible. This is to our knowledge, the first observation of a chiral-achiral metallic plasmonic interaction.

Three dimensional chiral structures such as metallic helices or NPs around DNA helix were found to exhibit strong CD effects in the visible. A major focus of this thesis work was the development of wafer-scale, three dimensional metal-decorated helical substrates with one of the largest reported optical responses in the visible. Additionally we investigated theoretically and experimentally the effect of plasmon coupling between the metal helices on the resultant CD and asymmetry factor. The effect of inter-particle separation was found to have a near-exponential dependence on the magnitude of the CD response. On the other hand, changing the refractive index of the dielectric template altered the chiral responses drastically.

Finally we investigated a novel geometry of chiral nanoshells consisting of a dielectric helical core with a conformal coating of a metallic shell. The spherical nanoshells have been extensively studied for its distinct plasmonic response and have been utilized for drug-delivery and optical sensing applications. Chiral nanoshells are fundamentally different because of the asymmetric nature of the nanoshell. Moreover the shell is made of alternate plasmonic material-Titanium Nitride which is optically similar to Gold but more robust and chemically stable in comparison. The resulting optical response of the chiral shell geometry was the broadest CD curve we observed until now, covering the whole of visible to near infra-red regime, implying this geometry to be a promising candidate for broadband circular polarizer applications.

All the studies carried out in this thesis, gives us an outlook on the possible design scheme and the underlying physics that could help us in engineering the chiral response based on the desired operating range of wavelength.