DOCTORAL THESIS

Numerical and analytical studies of ciricular dichroism of plasmonic nanospirals generated by glancing angle deposition

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Abstract

As emerging chiral metamaterials, plasmonic nanospirals (NSs) show strong optical activity that is expected to enhance the enantiodiscrimination of chiral molecules or help in the design of a new generation of integrated optical devices. The study of the optical activity of plasmonic NSs is still in its infancy, and no analytical model exists to describe their chiroptical mechanism. In this study, numerical and analytical simulations are devised to investigate the optical activity of plasmonic NSs that are generated by glancing-angle deposition. The findings will pave the way for the development of novel optical and optoelectronic devices with integrated functions.

The finite-element method is applied to numerically simulate the UV–visible light circular dichroism (CD) spectra of individual silver NSs (AgNSs), and the numerical results show good agreement with the CD spectrum of dispersed AgNSs. The optical chirality of the AgNS surfaces is numerically simulated to semiquantitatively account for the enantiomeric excess of photocyclodimerization of 2-anthracenecarboxylate induced by chiral nanoplasmons.

The CD spectrum of a closely packed random AgNS array has two CD peaks in the UV and visible regions with opposite signs. The pitch-normalized CD in the UV regime tends to be independent of the helical pitch, but that in the visible regime decreases in amplitude as helical pitch increases. The difference can be explained using an analytical LC circuit model and finite-element method simulation. The LC circuit model is used to quantitatively evaluate the chiroptical contribution. It is revealed that radiative loss makes an important chiroptical contribution to the two CD modes and that the visible CD mode receives a greater contribution from

radiative loss than does the UV CD mode.

Finally, the heterochiral biaxial AgNS arrays alter the sign of the visible CD by switching the incident direction, which shows that the arrays can function as circular polarizers in the visible regime. Furthermore, when AgNSs are deposited on a polymer substrate coated with indium tin oxide, the chiroptical flexible thin film has excellent chiroptical stability when exposed to forward mechanical bending, paving the way for the development of flexible or wearable chiroplasmonic devices.

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