

## Tunable plasmonic metamolecules and metasurfaces through bottom-up self-assembly

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DNA origami can be used to template assemblies of plasmonic nanorods, which can interact strongly with light, concentrating far-field radiation into sub-wavelength volumes. Predominantly, gold nanorods are used in such assemblies which interact with light in the NIR region, thus limiting their spectral response. We reported a method to grow silver shells of controllable thickness on gold nanorods and used them to create chiral assemblies using DNA origami, thus gaining control over the chiral and spectral response in the visible region.<sup>1</sup> Assemblies like these utilizing a variety of particles can achieve tailored optical functions, ranging from tunable chirality over SERS to fluorescence enhancement. However, applications are usually limited to those compatible with liquid phase colloidal suspensions. When deposited on surfaces, stochastic deposition places limits on usable metamolecules due to random aggregation and low surface density. Gopinath et al. reported a method combining nanosphere lithography and DNA origami placement<sup>2</sup> to create dense arrays on glass. We combine this method with our plasmonic metamolecules to explore the possibility of creating metasurfaces with a programmed optical response, chirality in our case. In the future, we envision expanding this fabrication method to build active metasurfaces with a dynamic response for biosensing and optical circuits for nanophotonic applications.

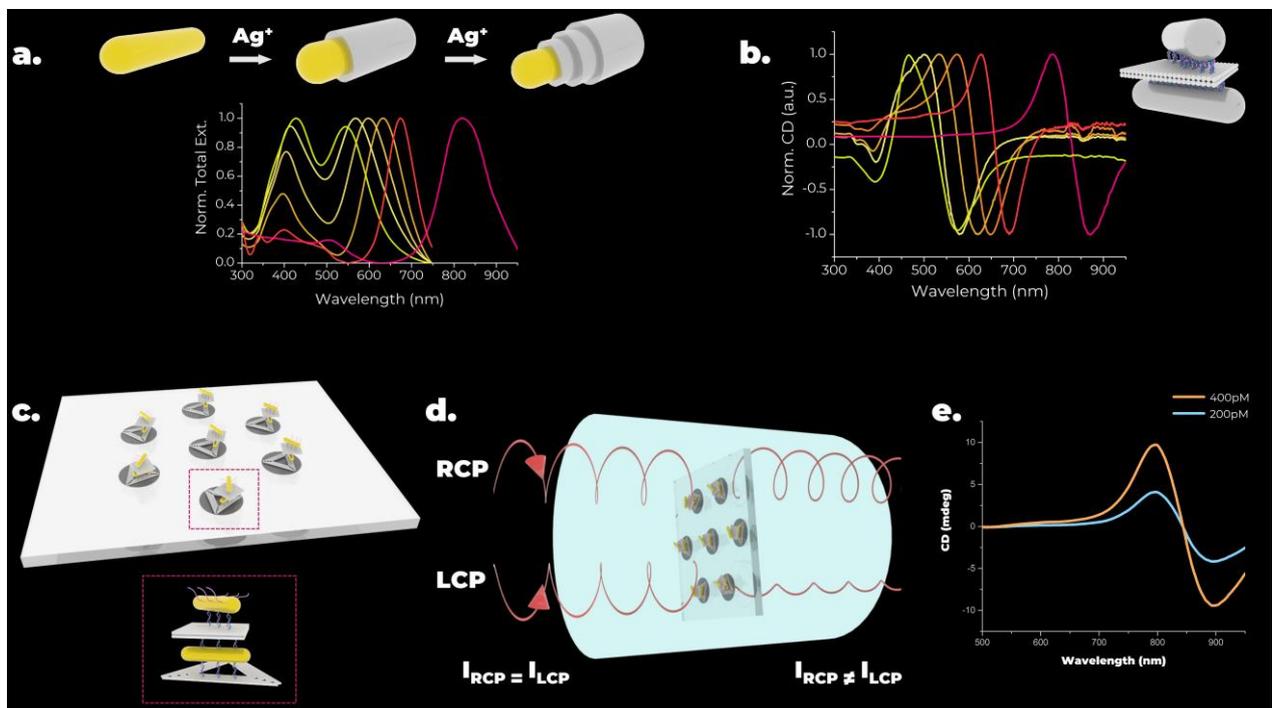


Fig. 1 Gold/Silver core/shell nanorods with (a) tunable extinction and (b) Circular dichroism (CD) over the visible spectrum. (c) Metasurface constructed with chiral metamolecules interact differently with circularly polarized right of different handedness (d), resulting in a bisignate CD spectrum.

1. Dass, M., Kuen, L., Posnjak, G., Burger, S., Liedl, T. Visible wavelength spectral tuning of absorption and circular dichroism of DNA-assembled Au/Ag core-shell nanorod assemblies (Accepted) Material Advances 2022.
2. Shetty, R. M.; Brady, S. R.; Rothmund, P. W. K.; Hariadi, R. F.; Gopinath, A. Bench-Top Fabrication of Single-Molecule Nanoarrays by DNA Origami Placement. ACS Nano 2021, DOI: 10.1021/acsnano.1c01150