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A bottom-up approach to the fabrication of optical metamaterials utilizing the self-assembly of silver dodecahedral clusters

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Abstract

In the production of optical metamaterials a key requirement on the comprising ‘meta-atoms’ is a light-induced magnetic response of similar magnitude to the induced electric response. It is desirable to achieve the assembly of such materials through bottom-up approaches, which require the production of colloidal dispersed ‘meta-atoms’. However, many of the currently realizable candidate colloidal particles present too weaker magnetic response for them to be utilized in this role. Hence, there is a need to for novel artificial meta-atoms with increased magnetic polarizability.

Previously, we presented our synthetic approach to the creation of suitable ‘meta-atoms’ in the form of a silica core decorated by 12 gold satellite particles (gold dodecapods, Au DDPs). We demonstrated that the optical magnetism of these objects arises from tangential polarization currents. The induced magnetic moment is analogous to that produced by circulating eddy (Foucault) currents in metals. The regular organization of the satellites around the core is more efficient than a random distribution of satellites for the generation of a circular current mode. Optical characterisation of such particles has shown them to be strong candidates for use as Huygens sources, and potential building blocks for the assembly of metamaterials.

We have subsequently improved the magnetic response of these particles through replacement of the gold with silver, [1] lowering optical losses by using a metal with a significantly higher plasmonic quality factor. [2] In this talk, we will present these improvements to the synthesis of DDPs alongside our ongoing work to build assemblies of these particles into real world metamaterials through bottom-up approaches.

Introduction

Optical metamaterials are ensembles of nanoresonators that exhibit unusual optical properties, otherwise unattainable in natural materials. One such property, a negative effective refractive index, first explored theoretically by Veselago in 1968, [3] was famously

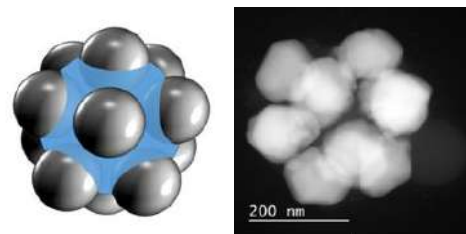


Figure 1 – Sketch of a silver dodecapod (12 Ag satellites arranged symmetrically around a dielectric core) and a TEM micrograph of a silver dodecapod.

experimentally realized in 1999 by Smith *et al*, who produced a composite medium possessing both a negative permittivity and permeability (i.e. a negative refractive index material). [4] Since this achievement, a wide variety of novel metamaterials have been developed, enabling the production of extraordinary optical devices such as invisibility cloaks and hyperlenses. [5,6] The range of applications of metamaterials has grown to include media enabling control of the propagation of acoustic, mechanical, thermal and seismic waves.

Until now, most optical metamaterials have been fabricated using top-down lithographic approaches, typically limiting the area of such metamaterials to below 100 μm^2 . Bottom-up approaches negate this problem, allowing the synthesis of nanoresonators and the fabrication of the metamaterial to be completed as separate steps, referred to as hierarchical self-assembly. [7] This approach allows complex nanoparticles with desirable electrical and magnetic properties to be synthesized as colloidal dispersions, and then assembled into structures on scales unachievable via top-down approaches.

Previously, our group has shown that assemblies of particles consisting of a dielectric core decorated with a disordered monolayer containing a large number of plasmonic nanoparticles (plasmonic raspberries) present a non-natural magnetic permeability. [8] We also demonstrated that well-controlled geometrically ordered

arrangements of 12 gold satellites significantly increases the obtainable magnetic dipole. [9] This magnetic moment could be increased further by using a metal with lower optical losses such as Ag in the satellites.

Taking this work forward, we are now seek to create metamaterials from our DDPs, which are extremely promising candidates.

Results

Highly symmetrical clusters were prepared via a multistep synthesis from dodecahedral-dimpled silica particles. The details of the synthesis are soon-to-be published [1], they are similar to our previously reported protocol utilizing Au [9]. The twelve silver satellites are arranged symmetrically around a 160 nm silica core (fig. 1).

The optically induced magnetic response of the Ag DDPs was measured by polarization-resolved static light scattering spectroscopy (fig. 2) and single particle scatter spectroscopy. The ratio of the magnetic to electric scattering is increased by a factor of ~ 6 compared to that of Au-DDPs with the same amount of metal. These results demonstrate the significantly reduced optical losses seen in silver. Hence, Ag DDPs are good candidates as Huygens sources for use in metamaterials.

This increase in the magnetic response is of a similar magnitude to that observed when replacing Au with Ag on our previously reported plasmonic raspberries [8]. However, the more uniform separation of the plasmonic satellites of the Ag DDPs, means that they possess a much greater response (around $6\times$ higher than the Ag raspberries).

Numerical simulations suggest monolayers of these particles will demonstrate near-perfect absorbance at ~ 400 nm. Current work focuses on self-assembling the Ag DDPs into colloidal monolayers through convective-assembly. The results of these experiments will be presented alongside their optical characterization.

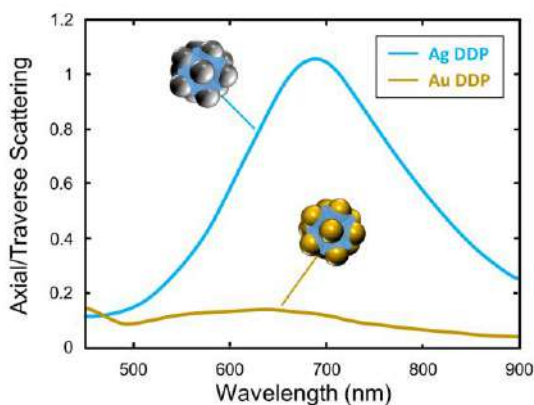


Figure 2: Ratio of axial scattering to transverse scattering, as obtained from static light scattering measurement. Ag DDPs show a significantly enhanced magnetic response (\propto axial scatter) compared with Au DDPs.

Conclusions

Bottom-up approaches offer a route to realizing large-area metasurfaces. Allowing the synthesis of highly promising candidate meta-atoms in bulk. Optimization of the particle characteristics by using Ag in the plasmonic satellites has enabled a significant increase in the induced magnetic response. Numerical calculations suggest assembling these particles into a 2D-array will yield a metasurface capable of near-perfect absorbance. Self-assembly methods will be employed to create Ag DDP monolayers to experimentally realize this result. This bottom-up approach will enable cost-effective, larger scale production of 2D and 3D metamaterials. [10]

Acknowledgements

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