STEM-EELS Studies of the Influence of Structural Imperfections in Plasmonic Metamaterials

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Controlled surface plasmon resonances are at the heart of an increasing variety of proposed technologies, ranging from telecommunications and data-storage to bio-sensing. In each case, the shape and position of metallic nanostructures are crafted to produce tailored plasmonic responses in both energy and spatial distribution. However, limitations in patterning and intrinsic materials properties including granularity and impurities limit the perfection to which the nanostructures can be fabricated. Here, we use a combination of simulations and Scanning Transmission Electron Microscopy with Electron Energy Loss Spectroscopy (STEM-EELS) to probe the profound impact such imperfections have on the plasmonic response.

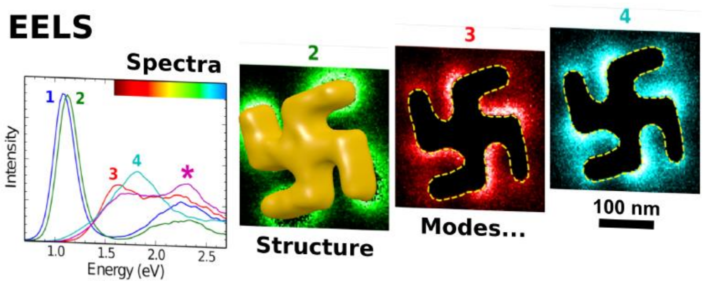

Figure 1. Deconvolution of STEM-EELS low-loss spectra reveal four distinct plasmonic excitation modes, with distinct spatial distributions that can be analysed through comparison with eigenmode simulations.

We focus on two specific systems: patterned chiral particles that are prototypical sensors of supramolecular structure; and near-field transducers with applications in hard disk drive data storage technologies. In both cases, the plasmons are supported by lithographically patterned gold nanostructures deposited onto SiN membranes; in both cases we are able to correlate deviations from the ideal optical spectra to nanometric fabrication imperfections. We use STEM images to provide an accurate survey of imperfect nanoparticle structure, from which we reconstruct a realistic 3-dimensional model for subsequent simulation. EELS spectra and plasmonic eigenmodes supported by the structures are then simulated within the MNPBEM Matlab toolbox [1,2]. In tandem, EELS data, collected on a JEOL ARM cFEG instrument, were analysed within the HyperSpy package [3] using Richardson-Lucy deconvolution to sharpen the energy resolution to 0.17eV. Typical results are summarised in Fig. 1, which displays the measured, deconvolved spectra from a chiral gold 'gammadion' shape that has biosensing applications [4]. Simulations indicate that the lowest order modes are expected to be (unusually) quadrupolar, then dipolar in nature, yet the experiment reveals all modes to have some optically-bright (dipole) character [5]. We find that even subwavelength-scale imperfections are sufficient to lift mode degeneracies, reduce the plasmon symmetry and confer dipole character on modes that are predicted to be dark. As a consequence, the mode energy, near-field magnitude and chiralty are modulated, with a consequent impact on the device functionality. In these structures, we show that coupling between plasmons on successive gammadian 'arms' can account for stabilisation of the quadrupolar mode and we use a simple hybridisation model to describe the resultant charge distributions, illustrated in the figure. The results will have broad applicability to a range of plasmonic devices and it is now clear that accurate device design must incorporate realistic structural and chemical imperfections.

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