Probing classical and novel optical coupling mechanisms by combined electron lithography and fast monochromated STEM-EELS

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The optimization of plasmonic structures that exploit coupled excitations for functional optical and electro-optical nanodevices, for applications such as bio-sensing, depends on a detailed understanding of how nanostructure material, size and geometry influence their optical response. Here we present a "tool box" for studying planar plasmonic structures which applies a fast, systematic approach consisting of two parts. Firstly, a wafer-scale preparation that uses a sequence of precise electron lithographic steps to fabricate TEM-ready samples with 1000s of reproducibly etched plasmonic nanoparticles or apertures, separated by defined gaps down to a few nm in size [1]. Secondly, their metrological analysis using fast (1200 spectra/s), high spatial statistics (e.g. 800 x 800 px) STEM-EELS hyperspectral mapping with an electron beam of good energy resolution (100 - 120 meV FWHM) and high beam current (220 - 250 pA), as carried out on a monochromated FEI Titan Themis 60-300 equipped with a Gatan GIF Quantum ERS spectrometer.

We first establish the metrological precision of the nanophotonic STEM-EELS mapping using classical plasmonic structures, namely dimer pairs of colloidal Au nanorods precisely arranged in lithographically-etched traps [2], and dolmens prepared from both the same colloidal nanorods and by lithographic etching [3]. We next present analysis of heterostructures, where we exploit the capability of carefully aligned electron lithography steps to create heterodimer nanoantennas consisting of Au-Ag and Au-Al nanodisc, nanorod, nanosquare or bowtie pairs (e.g. Fig. 1). We use the STEM-EELS mapping to explore the near field optical coupling of these metallic heterostructures, for instance identifying and explaining the excitation of different bonding and anti-bonding modes involving dipole and quadrupole excitations [4]. The results are interpreted with the help of an eigenmode analysis using a full wave method based on surface integral equations. By application of a novel antenna-based approach to interpret the EELS of nanostructures [5], we also unravel the energy lost to radiative scattering or each particle's relative absorption in longitudinally coupled nanorod pairs, seeing how they could be tuned for different applications.

With our experimental "tool box" giving the possibility to sample 40 - 50 unique structures in a day-long session, it can help lead to new insights for functional tailoring of plasmonic structures. Taking this forwards, as researchers look towards utilizing semiconductors for nanophotonics, we present ongoing work in which we apply our methodology to studying the near field optical excitations induced in Si nanoparticles of different geometries and coupling.

[1] Flauraud, V., et al., Nano Letters 17 (2017) 1703 - 1710
[2] Flauraud, V., et al., Nature Nanotechnology 12 (2017) 73 - 80
[3] Flauraud, V., et al., ACS Photonics 4 (2017) 1661 - 1668
[4] Flauraud, V., et al., ACS Nano 11 (2017) 3485 - 3495
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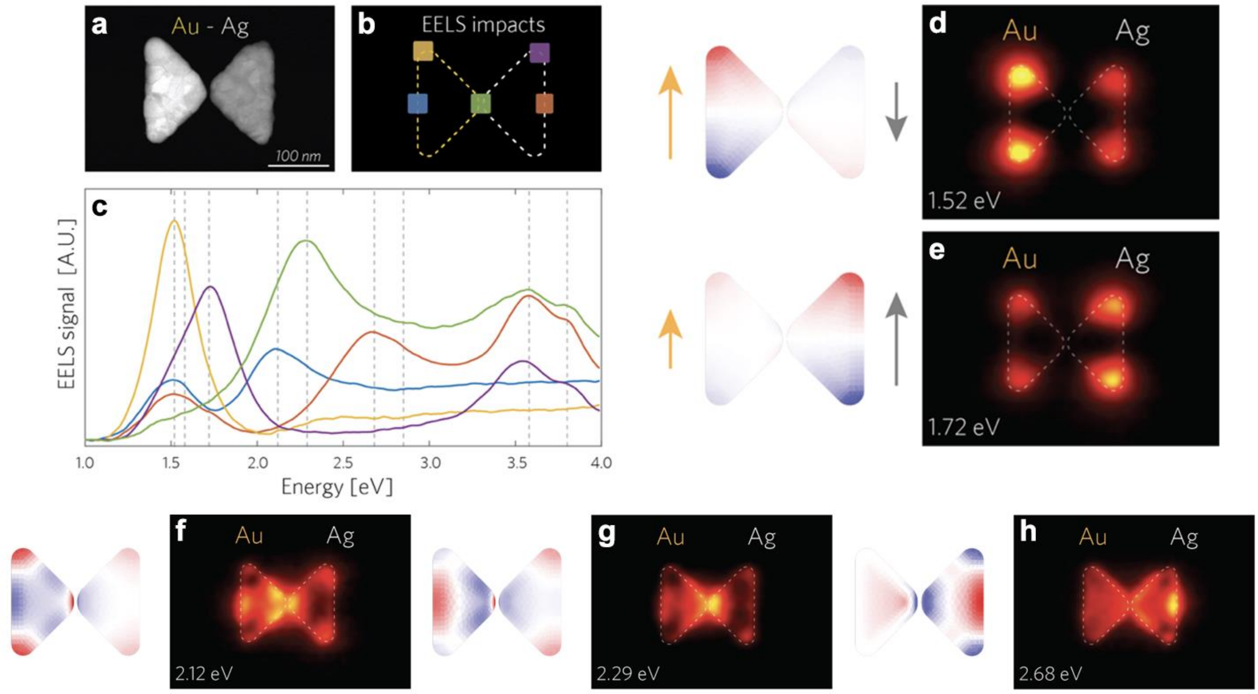

Figure 1. Coupling of Au:Ag bowtie nanoantenna showing: a) STEM image; c) EELS spectra integrated from different impact positions b); EELS intensity maps and corresponding computed eigencharge distributions for different plasmonic modes, from lowest energy transverse modes d) and e), to more complex higher energy couplings in f)-h).