## Mapping of Electron-Beam-Excited Plasmon Modes in Lithographically-Defined Gold Nanostructures

A.L. Koh,\*§ A.I. Fernandez-Dominguez,\*\* S.A. Maier,\*\* J.K.W. Yang,\*\*\* and D.W. McComb\*

- \* Department of Materials, Imperial College London, London SW7 2AZ, United Kingdom
- \*\* Department of Physics, Imperial College London, London SW7 2AZ, United Kingdom
- \*\*\* Institute of Materials Research and Engineering, A\*STAR, Singapore 117602

Electron energy-loss spectroscopy (EELS) can be used image localized surface-plasmon modes in metal nanostructures with unrivalled spatial resolution. By exciting plasmons with energetic electrons instead of far-field optical illumination, EELS also enables the efficient excitation of dark modes, which cannot be easily probed by optical means. [1]

Much of the work reported to date has been performed using chemically-synthesized nanoparticles randomly dispersed onto membranes. [2] This approach provides little control of particle arrangement and interparticle separations, thus limiting the complexity of plasmonic nanostructures that can be studied. In this contribution we use lithographically patterned plasmonic structures on thin membranes in order to experimentally map their plasmon modes. [3] This combination of high-resolution patterning and plasmon mapping is promising in lending direct experimental verification to theoretical predictions of plasmonic phenomena in complex nanostructures.

We have patterned polycrystalline gold nanostructures consisting of groups of triangles and rings and have systematically varied the distance between the nanostructures until they were connected in the middle. (Figure 1). EELS experiments were performed using an FEI Titan (S)TEM operated at 300 kV fitted with a monochromator and a Gatan Tridiem 866 spectrometer. The spatial and energy resolutions of the microscope in monochromated STEM mode are 0.3 nm and 0.2 eV (defined as the FWHM of the zero-loss peak), respectively.

The EEL spectra acquisition in the spectrum image (SI) mode presented information about the region of interest in the form of an "x-y-energy" data cube, where the first two axes corresponded to the lateral position of the specimen, and the third axis was the energy-loss spectrum. Using this mode, spectral information of plasmon resonances was obtained at every point in the region of interest, which can then be analyzed at a single point in space, or as a single energy slice across the entire region of interest (Figure 2). The latter approach allows us to clearly show the dependence of the corner, edge and coupled modes on the distance between the fabricated nanostructures, and to correlate these observations with theoretical predictions. [4]

The high resolution patterning method combined with high-resolution plasmon mapping of electron-beam excited plasmons allows investigation of plasmon resonances for a range of nanostructures and could have potential for use in systematic design verification of plasmon modes in arbitrary metal nanostructures. Furthermore, the high-resolution nature of the lithography and measurement techniques described suggests that this capability could be used to investigate plasmonic effects in sub-5-nm dimensions where quantum-mechanical effects become relevant [5, 6].

## References

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- § Now at, Stanford Nanocharacterization Laboratory, Stanford University, Stanford CA 94305, USA

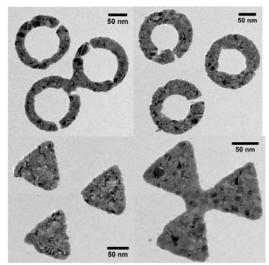


Figure 1 : TEM images of some of the polycrystalline Au nanostructures prepared by e-beam lithography.

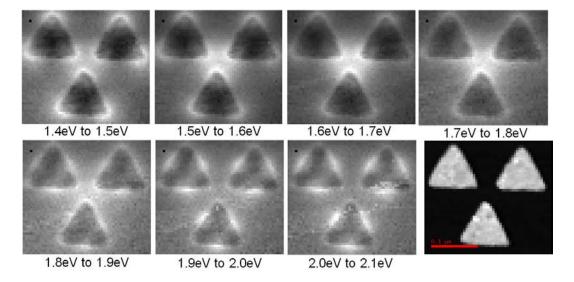


Figure 2 : Energy slices ( $\Delta E = 1 \text{eV}$ ) extracted from the EELS spectrum image (bottom right) recorded from one of the nanostructures formed by e-beam lithography