

## Near-Field Mid-Infrared Plasmonics in Complex Nanostructures with Monochromated Electron Energy Loss Spectroscopy

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Localized surface plasmon resonances (LSPRs) in complex nanostructures have seen widespread use in bringing optical phenomena to on-chip applications [1]. For dipolar excitations in symmetric nanoparticles of electron dense materials like Au, Ag, or Al, LSPRs are usually in the visible range. However, in complex nanostructures composed of those materials, unique IR and telecom plasmon resonances can emerge [2].

The nanoscale confinement of light to the surface of a structure makes LSPRs highly versatile in nanotechnology, however the confinement also makes them difficult to study at the near-field through traditional optical methods due to the diffraction limit. Scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS) can be used to map plasmon resonances directly at the nanoscale, but the inherent energy spread of the electron probe has long made plasmon modes in the IR spectrum difficult to observe. However, recent advances in monochromation have greatly increased the energy resolution of STEM-EELS and greatly reduced the background in the IR [3].

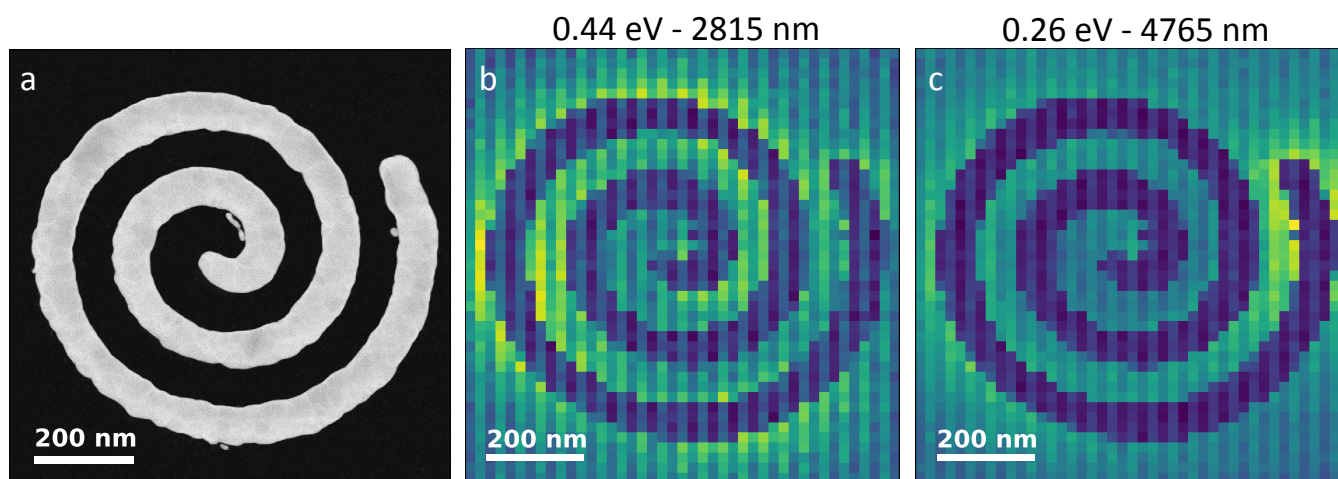
Here we examine Au Archimedean nanospirals. The structures have strong potential for on-chip applications due to the highly-tunable non-centrosymmetric geometry that supports many unique plasmon modes, as well as producing many interesting optical phenomena such as second harmonic generation and super-continuum luminescence [4,5]. The plasmon modes extend across a broad spectral range and many are primarily active in the IR, making the structure ideal for monochromated EELS.

Figure 1a shows a high angle annular dark field (HAADF) image of a Au nanospiral with a winding number of  $5\pi$ , arm width and spacing of 80 nm, and a Au film thickness of 40 nm. A spectrum image (SI) of the nanospiral is acquired with a Nion monochromated, aberration-corrected UltraSTEM operated at 60 kV [6]. Monochromation is performed by using a variable slit to balance the energy resolution with the probe current. To maximize signal-to-noise while minimizing the energy spread in the probe, the slit is adjusted so that the zero loss peak has a full-width half-maximum of 75 meV.

Two distinct peaks are observed in the IR range from the SI, at 440 meV and at 260 meV, which corresponds to wavelengths of 2815 nm and 4765 nm respectively, and each exhibiting a completely different spatial profile. The ability to map complex mid-IR plasmon modes with a monochromated aberration corrected STEM opens new avenues to explore on-chip optical technologies with sub-Ångstrom spatial resolution [7].

## References:

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**Figure 1.** (a) HAADF-STEM image of the Archimedean Au nanospiral. (b) and (c) Spectrum images of distinct plasmon modes in the Mid-IR, at 2815 nm (b) and 4765 nm (c)