Multipole plasmons and their disappearance in few-nanometre silver nanoparticles - DTU Orbit (12/11/2019)

**Multipole plasmons and their disappearance in few-nanometre silver nanoparticles**

Electron energy-loss spectroscopy can be used for detailed spatial and spectral characterization of optical excitations in metal nanoparticles. In previous electron energy-loss experiments on silver nanoparticles with radii smaller than 20 nm, only the dipolar surface plasmon resonance was assumed to play a role. Here, applying electron energy-loss spectroscopy to individual silver nanoparticles encapsulated in silicon nitride, we observe besides the usual dipole resonance an additional surface plasmon resonance corresponding to higher angular momenta for nanoparticle radii as small as 4 nm. We study the radius and electron beam impact position dependence of both resonances separately. For particles smaller than 4 nm in radius the higher-order surface plasmon mode disappears, in agreement with generalized non-local optical response theory, while the dipole resonance blueshift exceeds our theoretical predictions. Unlike in optical spectra, multipole surface plasmons are important in electron energy-loss spectra even of ultrasmall metallic nanoparticles.

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