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Probing plasmon resonance’s dependence on gap size in silver dimers by EELS

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Improvement in the energy resolution of modern analytical transmission electron microscopes (TEMs) has enabled electron energy-loss spectroscopy (EELS) in the visible light energy range and below. Aggregates of multiple silver nanoparticles, in which coupling of the particles results in highly confined and enhanced local fields in the nanometre size gaps between them, are of particular interest for various applications, including surface enhanced Raman spectroscopy [1]. While, most optical techniques do not hold the spatial resolution to image such small dimensions, EELS combined with scanning TEM (STEM) can probe Ångström-scale dimensions. We have studied silver dimers, as the simplest multiparticle plasmonic structure, with EELS. Changes of the dipolar plasmon resonances in EELS spectra of dimers as a function of the interparticle distance are monitored. Experimentally observed shifts of plasmon resonance are compared with computations, using a multiple-scattering simulation formalism [2]. The measurements and calculations were carried out for spherical particles ~20 – 30 nm in diameter.

Plasmon resonance’s dependence on gap size in silver dimers

- Redshift of the dipolar plasmon with gap size in EELS from silver spheres of diameters 20 – 25 nm.
- The dipolar plasmon redshift ratio \( (E_0^{\text{dip}} - E_0^{\text{mono}})/(E_0^{\text{dip}}) \) of silver dimers of 20 – 30 nm diameter, as a function of the diameter to the centre to centre distance ratio \((2R/(d+2R))\).
- EELS measurements are compared with simulations of optically excited dipolar plasmons of silver dimers, using a multiple scattering formalism based on the Lippmann-Schwinger equation and the electromagnetic Green’s tensor [2]. A linear fit for the logarithmic plot of the measurements can be considered (giving a scaling to the power 6.6 of plasmon energy with gap size) but the plots indicate a stronger redshift in dipolar plasmon energy for smaller gaps (below ~ 2 nm surface to surface gap here).

Conclusions

- High energy-resolution EELS is a powerful technique for probing the plasmonic properties of nanostructures with a high spatial resolution.
- The scaling of the dipolar plasmon resonance in silver dimers was investigated with EELS and computations. Experimental measurements are in good agreement with electromagnetic calculations for gaps down to ~1 nm.

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References