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The nano-gap and the emitting molecule: Control of polarization and spectral shape

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The realization of single-molecule surface-enhanced Raman scattering (SERS) from molecules positioned within nano-gaps between metallic nanopraticles has opened up exciting opportunities for studying plasmonic fields and their effects on quantum emitters. We recently showed that constructs made of pairs of nanoparticles with an individual molecules bridging their gap can be systematically formed and studied [1]. By changing the size of the particles, we were able to tune the position of the plasmon resonance spectrum, so that the overlap with different parts of the molecular Raman spectrum changed, leading to significant modulation of its shape. More intricate control over molecular properties can be achieved if a third particle is added to the contstruct. It was found that by breaking the dimer symmetry, a third particle can couple strongly to the emitted Raman field and modulate its polarization in a wavelength-dependent fashion [2]. This surprising experimental result was backed up by a series of Generalized Mie calculations, showing the effect of the distance of the third particle, its size and position [3]. Interestingly, the refractive index of the surrounding medium serves as another control parameter that allows changing the coupling between the particles and modulating the polarization of emitted light.

[1] Dadosh T, et al. (2009) Plasmonic Control of the Shape of the Raman Spectrum of a Single Molecule in a Silver Nanoparticle Dimer. Acs Nano 3:1988-1994.
[2] Shegai T, et al. (2008) Managing light polarization via plasmon-molecule interactions within an asymmetric metal nanoparticle trimer. Proc Natl Acad Sci USA 105:16448-16453.
[3] Li ZP, Shegai T, Haran G, Xu HX (2009) Multiple-Particle Nanoantennas for Enormous Enhancement and Polarization Control of Light Emission. Acs Nano 3:637-642.