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Plasmonic nanoantennas with sub-nanometer gaps: fabrication and observation of quantum mechanical effects

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In surface-enhanced Raman scattering (SERS), the giant field enhancements generated by plasmonic nanostructures compensate for the relatively weak Raman scattering cross sections of many molecules. The effect is so substantial that Raman spectroscopy can be performed on single molecules (e.g. [1]). A structure that has proven especially effective for SERS is the dimer nanoantenna, i.e. two metal nanoparticles separated by a small gap.

In this presentation, works by the author and his team on advanced and novel nanofabrication method for plasmonic dimer nanonantennas with very small gaps will be described.

The first method involves the use of volume expansion that chromium undergoes upon oxidation. We demonstrated arrays of plasmonic nanoantennas, with gaps as narrow as 3 nm [2]. These devices were used in demonstrations of SERS substrates with very high enhancement factors (up to ~10^{10}, [3]) and for beamed Raman scattering [4].

In the second method, gold nanodisk dimer antennas were produced via a fabrication method employing two lithography steps. Each step defined one half of the antenna, i.e. one gold nanodisk, and careful alignment was performed. Using our method, gaps as narrow as ~2 Angstroms were achieved. We performed SERS on thiophenol monolayers formed on the antennas. We demonstrated that SERS enhancements increase as the gap width is reduced from 10 nm down to 6.7 Angstroms. As the gap reduced from 6.7 down to 2 Angstroms, SERS enhancements were seen to decrease. This was found to be consistent with a model that incorporated quantum mechanical tunnelling across the gap [5]. This work therefore demonstrated that quantum mechanical effects limit the field enhancement of plasmonics, and overturned the existing notion that smaller gaps were always favourable for SERS.

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