DNA Origami Used to Assemble Nanoplasmonic Structure

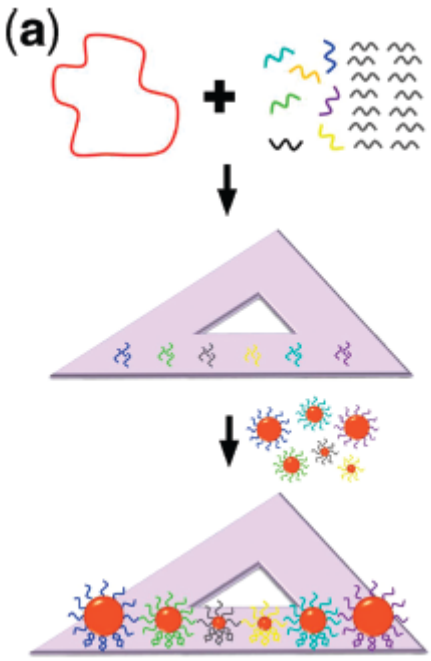
Ding et al. report on the use of deoxyribonucleic acid (DNA) origami as a scaffold for the assembly of gold nanoparticles into a carefully organized structure to create, ideally, a device with a strongly enhanced plasmonic resonance.

DNA origami has attracted a great deal of attention since it was first described (Rothemund, Nature (2006)). In brief, a long (7,000-base) single-strand loop of DNA derived from a virus (M13mp18) is mixed with approximately 200 short (typically 32-base) "staple" strands which, by hybridizing with the viral strand, cause it to fold into a structure determined by the binding locations of the staple strands. The size of the completed origami is roughly 100 nm in diameter. In addition to being an elegant approach to producing precisely engineered DNA structures, DNA origami has the potential to act as a platform for the molecularly precise arrangement of other nanoscale objects. This is because it is possible to extend "sticky-ended" DNA from the origami, enabling the attachment of complementarily DNA-functionalized objects.

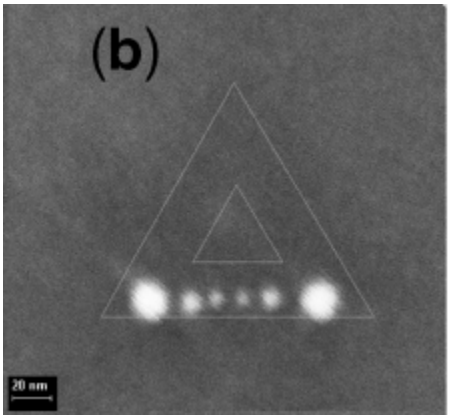
Ding et al. chose to realize a plasmonic nanolens structure first analyzed by Li et al. (Physical Review Letters (2003)). Those authors described how a chain of three self-similar gold nanoparticles can give rise to field enhancements of up to a thousand-fold, depending on the wavelength of light used and the precise arrangement of the nanoparticles. The greatest field enhancements are obtained when the smallest particle is on the order of 10 nm in diameter with particle-to-particle spacings as small as 9 nm. These dimensions are difficult to obtain reliably with lithographic methods, making an alternative approach to fabrication attractive.

In order to arrange the nanoparticles on the origami, the authors extended sticky ends from specific staple strands on the origami. The sticky ends were designed to be complementary to thiolated DNA strands attached to the gold nanoparticles with sizes of 5 nm, 10 nm and 15 nm. By choosing to functionalize the different sized gold particles with different DNA strands, each size of nanoparticle could be directed to the desired location on the origami. In this way, the authors were able to attach sets of six nanoparticles, arranged in order, to the origami. Interestingly, the authors found that three DNA linkages were needed to attach the nanoparticles successfully – control experiments using only two attachment strands lead to poor yields and inaccurate nanoparticle positioning.

Once they had produced origami with the plasmonic devices, the authors conducted preliminary measurements of their optical response, observing a 5 nm shift in the location of the plasmon-band peak in the ultraviolet-visible absorbance spectrum, showing some level of plasmonic interaction. One question that always arises in the context of trying to model and measure the plasmonic response of a nanostructure is: how sensitive is the response to the precise details of particle size, shape and spacing? This is particularly important given the difficulty of controlling the morphology of such small objects. Another key question is: how will the completed nanoparticle structures be integrated into larger-scale devices? Fortunately, this is an active area of research with recent papers by Kershner et al. and Hung et al. (Nature Nanotechnology (2009)) focusing on the use of lithographically-patterned, chemically-functionalized surfaces to control the placement and orientation of origami. Finally, and perhaps most critical, as far as applications directed towards integration into silicon electronics, is the question of what are the fundamental limits in terms of the overall yields that can be achieved?



Schematic of the assembly process showing the addition of single-stranded viral DNA and the staple strands, with some specially configured to provide sticky ends for nanoparticle attachment, to form a triangular origami tile with attached, DNA-functionalized Au nanoparticles.



Scanning electron microscope image of a single set of Au nanoparticles assembled onto a DNA origami template. The triangle shows the location of the origami relative to the nanoparticles.