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Plasmonic Enhancement of Raman Signal using Complex Metallic Nanostructures based on DNA Origami

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DNA-based nanostructures, such as "DNA origami," have recently emerged as one of the leading techniques for precise positioning of nanoscale materials in fields ranging from computer science to biomedical engineering. The origami is composed of a single scaffold DNA strand to which smaller "staple" strands are attached through DNA complementarity. The staples help to fold the scaffold strand into the designed structure of a predetermined shape. The resulting templates are highly addressable and have proven to be versatile tools for site-specific placement of various nanocomponents, such as metallic nanoparticles, quantum dots, fluorophores, etc. Building upon massively paralleled assembly mechanism of the origami and its ability to position nanocomponents, one may hope to utilize it for biosensing purposes. One attractive goal is the Raman spectroscopy, which provides a highly specific chemical fingerprint. Unfortunately, the Raman scattering cross section is small; Surface Enhanced Raman Spectroscopy (SERS) enhances the otherwise weak Raman signal by trapping the analyte molecules in the regions of intense electric field produced near rough metallic surfaces. These "hot spots" can be understood as resulting from localized surface plasmon modes resonantly exited by the incident laser excitation. We have earlier shown that metallic nanoparticles controllably attached to DNA origami can be further enlarged via an in-solution metallization; this technique allowed us to build metallic structures of complex topology. Recently, we have performed Raman spectroscopy of molecules attached to these metallic assemblies. Specifically, DNA origami is first used to organize the metallic structures, followed by a covalent attachment of Raman-active molecules to the metal. We found that the substrates with four nanoparticles per origami produce a strongly enhanced Raman signal compared to the control samples with only one nanoparticle per origami for the same particle density. Furthermore, in the samples with four particles per origami the Raman signal systematically decayed as a function of the laser exposure time. Similar behavior has been previously reported and attributed to photo-damaging effects of the high intensity fields at the "hot spots." In the samples with four nanoparticles per origami, the hot spots are located between the pairs of NPs; the one-particle control samples lacking the inter-particle hot spots showed no decay of the Raman signal, confirming our conclusion.