Title: Towards strongly-coupled emitter-metal hybrid nanosystems: Selective attachment of flurorescent dye molecules to Au crystals using DNA linkers

Authors: Tamiko Masuda, Jannu Casanova-Moreno, Jonathan Massey-Allard, Dan Bizzotto and Jeff F. Young

Quantum oscillators with large dipole transition moments can coherently couple with strongly localized photon states if the single photon Rabi frequency exceeds relevant decoherence rates. For a given two level system, its electromagnetic environment has to be engineered to maximize the cavity Q, and minimize the volume V of the localized photon mode, as $Q/\sqrt{V}$. The required conditions have been achieved in both macro- and micro-scopic dielectric cavities using, respectively, real and artificial atoms.

Although clever engineering of, for instance, photonic crystal microcavities in semiconductor slabs, can reduce the mode volume to below a cubic wavelength, the associated maximum electric field is still far less than that near the surface of nanoscale noble metal crystals at frequencies of localized plasmon resonances. Although the dephasing rates of these plasmon modes is large, theoretical predictions [1] of strong coupling of quantum dots with highly localized, dark plasmon modes, motivates the development of techniques for localizing quantum oscillators to specific locations on the surface of noble metal nanoparticles with nanometer precision.

Here we describe progress in this direction using electrochemical and electroless chemistry to selectively attach one end of 10 nm long DNA strands to the (111) surface of gold crystals with the other end functionalized with florescent dye molecules (Bodipy 650-665). Fluorescence imaging of macroscopic spherical gold crystals proves that protective surfactant molecules can be selectively desorbed from the (111) facets and then replaced with the DNA strands. The achieved electroless desorption process opens the door to selective functionalization of noble metal nanoparticles.

[1] C. Van Vlack, et. al., *Phys. Rev. B*, **85**, 075303 (2012)