Coupling fluorescent molecules to plasmonic antennas with DNA

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DNA self-assembly is a flexible and robust technique to produce hybrid nano-structures. Here, we use a short DNA double-strandto position organic dye molecules in the gap of gold nanoparticle dimers that act as antennas for light (Figure 1-a). These nanoantennascan enhance the spontaneous emission rates of dye molecules by more than two orders of magnitude [1-2]. However, the efficiency of the emitter-antenna interaction strongly depends on the size of the plasmonic particles. We have demonstrated that DNA-templated 60 and 80 nm diameter gold nanoparticle dimers, featuring one fluorescent molecule, provide single-photon emission with lifetimes that can fall below 10 ps and typicalquantum yields in a 45–70% range (Figure 1-b) [2].

Furthermore, the versatility of DNA allows the precise positioning of more than one fluorescent emitter in a plasmonic antenna. For instance, we have introduced two molecules with overlapping absorption and emission spectra, thus allowing non-radiative Förster resonant energy transfer (FRET) (Figure 1-c). This system allows us to highlight the competition between radiative and non-radiative decay channels in complex photonic nanostructures [3]. Finally, we will discuss the potential of DNA to incorporate a large number of fluorescent molecules in a plasmonic antenna in order to influence many-body interactions.



Figure 1:(a) DNA-linked gold particle dimers, associated with a single fluorescent molecule, which are freely diffusing in solution. (b) Fluorescence enhancement factors for different particle sizes, estimated in fluorescence correlation spectroscopy and compared to Mie theory. (c) Normalized fluorescence decay traces of a donor dye in the absence (empty markers) and presence (filled markers) of an acceptor.

^{1.} Busson M. P. et al., Accelerated single photon emission from dye molecule-driven nanoantennas assembled on DNA, Nat. Commun. 3, 1-6, 2012

^{2.} Bidault S. et al., *Picosecond Lifetimes with High Quantum Yields from Single-Photon-Emitting Colloidal Nanostructures at Room Temperature*, ACS Nano, DOI: 10.1021/acsnano.6b01729, 2016

^{3.} Bidault S. et al., *Competition between Förster resonance energy transfer and donor photodynamics in plasmonic dimer nanoantennas*, ACS Photonics, DOI: 10.1021/acsphotonics.6b00148, 2016