Manipulating the Excitation and Emission of Single Molecules coupled to Optical Antennas assembled with DNA Origami

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In this contribution, we self-assemble optical antennas based on Au and Ag colloidal nanoparticles with sizes up to 150 nm (see Fig. 1a) where single photon emitters such as organic fluorophores are placed at the hotspot. We show that these antennas can outperform top-down lithographic antennas in terms fluorescence intensity and photo-stability reaching a photon count rate enhancement of three orders of magnitude and a 30x enhancement in the average number of emitted photons [1-2]. Furthermore, these antennas can operate in a broad spectral range throughout the visible spectrum [3].

We also exploit these structures to study how optical antennas mediate the emission of single photon emitters. We perform two different experiments. In the first one, we use super-resolution techniques such as DNA-PAINT, to determine how the coupling between a single photon emitter and a single Au nanoparticle affects the apparent emission center (Fig. 1b) [4]. This approach allows us to quantitatively measure a shift in the emission origin, towards the nanoparticle center, of up to 30 nm. In the second set of experiments, we study how the fluorophore-antenna coupling affects the emission pattern. We compare the situation of a freely rotating fluorophore positioned within a DNA-origami structure with and without an optical antenna dimer (Fig. 1c). Back focal plane imaging and polarization dependant measurements show that the presence of the optical antenna mediates the emission and leads to a dipolar pattern determined by the dimer orientation.

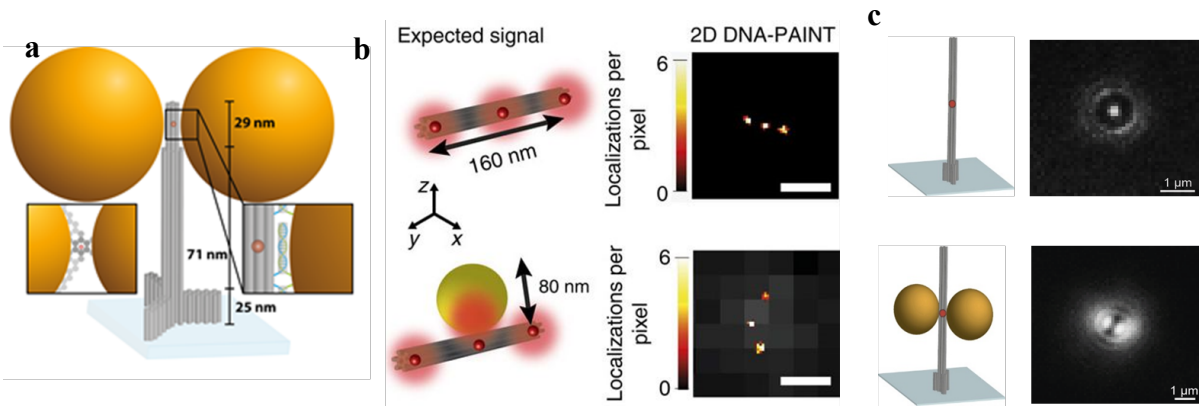

Fig. 1 (a) Sketch of an optical antenna dimer based on Au colloidal nanoparticles self-assembled onto a DNA-origami structure. A single fluorophore (in red) is positioned at the antenna hotspot. (b) Super-resolution measurements reveal that a single Au nanoparticle can shift the apparent emission center of single fluorophores. (c) Back focal plane measurements of a freely rotating fluorophore in the absence (upper) and presence (lower) of an optical antenna dimer.

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