DNA ORIGAMI SELF-ASSEMBLED HIGH-INDEX DIELECTRIC OPTICAL ANTENNAS FOR SINGLE-MOLECULE FLUORESCENCE MANIPULATION

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Abstract: We self-assemble optical antennas based on silicon nanoparticles using the DNA origami technique. We exploit these structures to manipulate the fluorescence properties of single fluorophores placed in their proximity. We focus on effects on the fluorescence lifetime, intensity and directivity.

Over the last decade, the DNA origami method[1] has established itself as one of the most versatile techniques for the bottom-up synthesis of hybrid species with tailored functionality[2]. This approach, which is based on the self-assembly of DNA strands into arbitrarily designed 3D structures, can be exploited to organize different entities, such as fluorophores, proteins, or NPs with nanometer precision and stoichiometric control. Consequently, it has been widely used to produce a variety of optical antennas for sensing and light manipulation at the single molecule level in controlled geometries. However, to date this approach has been constrained to Au, Ag and QD NPs with no works reported so far on combining DNA origami with high-index dielectric NPs. In this work, we develop a technique to functionalize high-index dielectric colloidal Si NPs[3] with DNA sequences using click-chemistry. To further demonstrate these findings, we self-assemble Si NP dimers using the DNA origami technique. Finally, we also exploit the DNA origami technique to position both Si NPs and organic fluorophores at controlled gaps to study the distance dependance energy transfer, the modification on the fluorescence rates (figure 1b) and the emission directivity. Our results show that in the vicinity of the Si NPs the radiate rate increases more than the non-radiative rate and that the Kerker condition can be exploited to obtain emission unidirectionality (figure 1c).

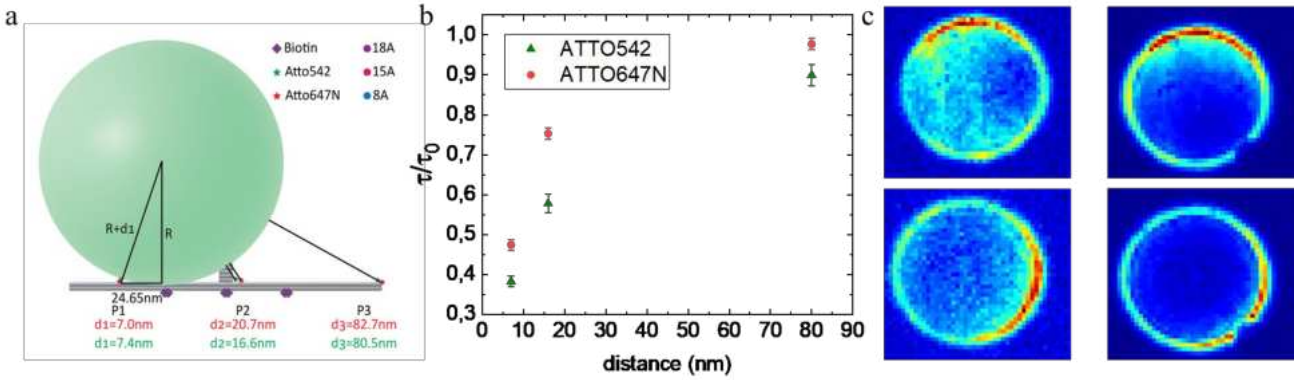

Figure 1. (a) Sketch of the DNA origami structure hosting a single Si NP and fluorophores at determined positions. (b) Fluorescence lifetime as a function of the distance to the nanoparticle. (c) Back-focal plane images

depicting the directivity induced by the presence of the Si NP.

References

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