Controlling the Radiative and Non-Radiative Decay Channels of Fluorescent Emitters with Plasmonic Nanoantennas

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By locally enhancing and confining electromagnetic fields at optical frequencies, plasmon-based optical antennas can increase the power dissipated by isolated fluorescent emitters but also influence their non-radiative coupling to nearby molecules. To control, at the nanoscale, the near-field interaction between a fluorescent molecule and an optical antenna, we use a short DNA double-strand to position individual organic dyes in the gap of gold nanoparticle dimers (Figure 1-a). These nanoantennas can enhance the spontaneous decay rates of quantum emitters by more than two orders of magnitude (Figure 1-b) [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 nm and 80 nm diameter gold nanoparticle dimers, featuring one fluorescent molecule, provide single-photon emission with lifetimes that can fall below 10 ps and typical quantum yields in a 45−70% range (Figure 1-c) [2].

Furthermore, the versatility of DNA allows us to position 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].

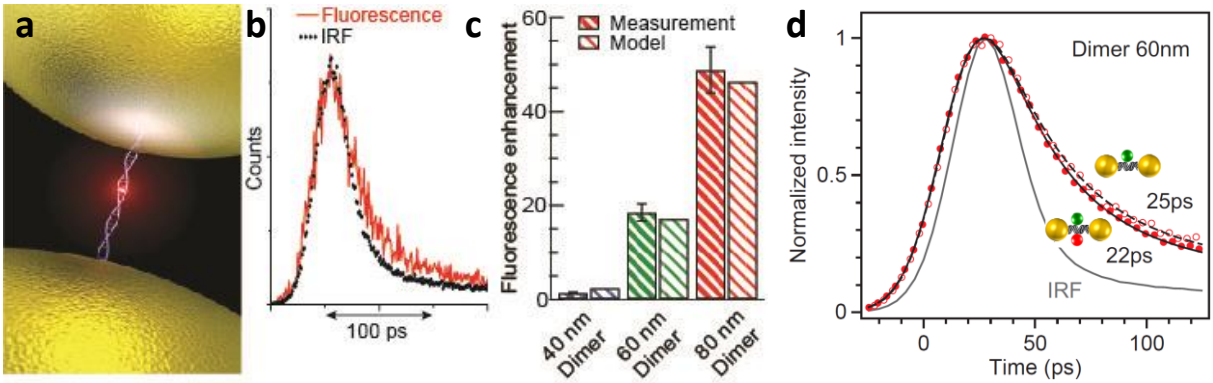

Figure 1: (a) Schematic representation of DNA-linked gold particle dimers, associated with a single fluorescent molecule. (b) Fluorescence decay trace of a particle dimer with a single emitter (fluorescence lifetime of 10 ± 5 ps). (c) Fluorescence enhancement factors for different particle sizes, estimated 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