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.

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