

DNA as a Smart Template to Strongly Couple Fluorescent Molecules with Plasmonic Nanoantennas

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DNA nanotechnology provides a flexible toolbox to produce hybrid nanostructures with a nanoscale control over the position and relative orientation of inorganic nanoparticles. In particular, this approach can be used to introduce a known number of dye molecules inside a plasmonic resonator made of two gold nanoparticles. In a weak electromagnetic coupling regime, DNA-templated gold particle dimers enhance the fluorescence rate of single molecules by more than two orders of magnitude, providing bright ultrafast single-photon sources with quantum yields above 50% [1]. However, it would be particularly attractive to increase the electromagnetic coupling strength in order to use such hybrid nanostructures as building blocks for quantum technologies at room temperature.

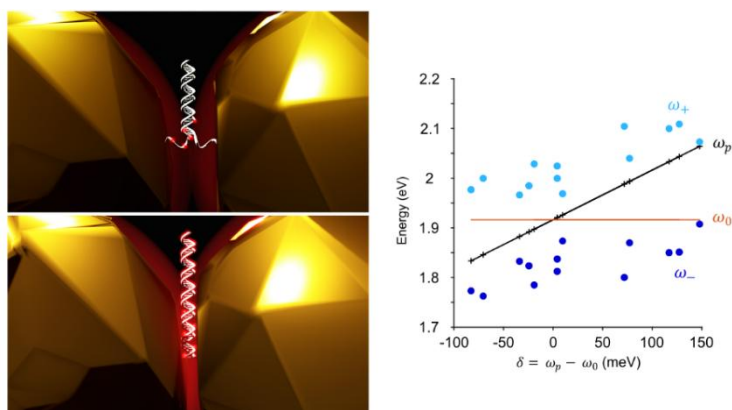


Figure 1. (a) Schematic representation of 5 dye molecules coupled to a dimer of gold nanoparticles with an interparticle distance that is actively reduced below 2 nm by screening electrostatic repulsion between particles. (b) Distribution of hybrid wavelengths ($\omega_{+/-}$) compared to the resonance wavelength of the molecules (ω_0) and the estimated longitudinal plasmon resonance wavelength (ω_p).

To produce hybrid nanostructures, in which a known number of dye molecules are strongly coupled to a plasmonic resonator, we produce 40 nm gold dimers featuring 5 ATTO647N molecules and we actively decrease the interparticle distance below 2 nm (Fig. 1-a). Using scattering spectroscopy, we observe plasmon mode splitting in the longitudinal resonance of single 40 nm gold particles only when the spacing is below 2 nm [2]. The wavelength distribution of hybrid eigenmodes features an anticrossing behavior, typical of a strong-coupling regime, in excellent agreement with electrodynamic simulations (Fig. 1-b). Furthermore, we demonstrate the influence of the planar facets of polycrystalline gold particles on the probability of observing this coupling regime, highlighting which geometrical parameters must be further optimized to reach single-molecule strong coupling at room temperature in a controlled and reproducible way.

[1] S. Bidault et al, ACS Nano 10, 4806–4815 (2016)

[2] J. Heintz et al., submitted (21)