DNA Origami-Templated Assembly of Heterogeneous Nanocavity for Quantum Emitter

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Quantum optics studies the interaction between light and matter where single quanta of light (photons) and single entities of matter (quantum emitters) are controllably coupled.[1] As the coupling strength between the emitter and the nanocavity increases, profound effects emerges: mixed states are produced that are part light, part matter. The ability of nanostructured metamaterials to tailor and control measurable changes in the quantum properties of light has resulted in a breakthrough that could play a prominent role in the future of computing by using photon spin and orbital momentum energy transfer to encode quantum information. However, serious challenges still exist in the stringent design and manufacturing of an efficient quantum interface that enables strong interactions between an optical nanocavity and a single quantum emitter.

DNA nanotechnology has been known as a versatile nanofabrication tool.[2] Here, we devised a strategy to construct plasmonic nanocavities that contain an ultra-small gap by using DNA origami (DO) guided gold nanorod (AuNR) assembly. Three-dimensional DO containing tubular grooves were synthesized to precisely align two AuNRs in a linear configuration (Figure 1). A saddle-shaped spacer was placed between AuNRs to adjust their gaps, and most importantly, deterministically introduce a single fluorescent emitter at nanometer accuracy within the cavity. This assembly strategy is advantageous in that anisotropic nanoparticles could be organized in a tip-to-tip fashion, which greatly reduces the cavity volume and enhances light-matter coupling. By carefully designing the the spacer and AuNR docking strands, precisely confined gaps of only 2-3 nm were achieved, which were highly desired for strong coupling studies (Figure 2). The as-prepared AuNR-DO assembly was examined under spectrometer and microscope, revealing scattering peaks corresponding to the emitter and the AuNRs. Additionally, a 20-30 fold enhancement in the fluorescent emission was recorded, which was accompanied by a significant decrease of emission lifetime from 1.9 ns to 42 ps (Figure 3). The above discoveries clearly demonstrate the capability of DNA-templated assembly of heterogeneous nanocavities to study quantum-scale strong light-matter interaction.

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[1] M. S. Tame, K. R. McEnery, Ş. K. Özdemir, J. Lee, S. A. Maier and M. S. Kim, Nature Physics **9**, 329 (2013).
[2] P. W. K. Rothemund, Nature **440**, 297 (2006).

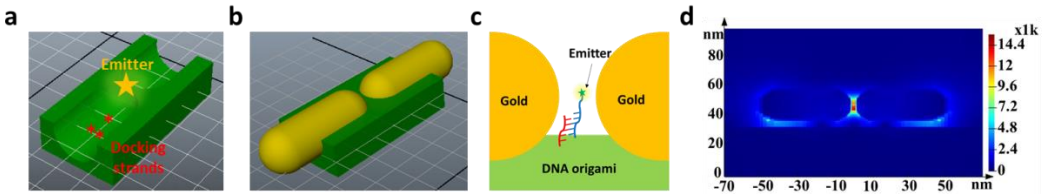

Figure 1: Scheme of the DNA-origami assembled nanocavity-emitter design. (a) Scheme of the DNA origami. The position of emitter and half of the docking strands are labelled. (b) Scheme of the AuNR-DO assembly. (c) Schematic view of the single emitter in the nanocavity. (d) Optical simulation showing the local field enhancement.

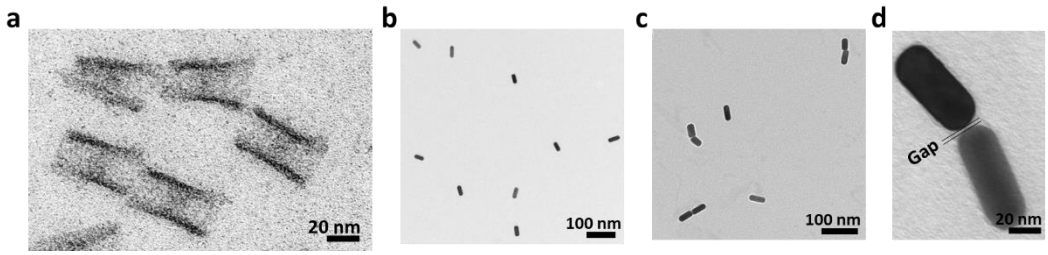

Figure 2: DNA origami templated AuNR assembly. (a) TEM image of the DNA origami synthesized in this work. (b) TEM image of the AuNR solution. (c) TEM image of the AuNR assembly after annealing with DO. (d) A magnified TEM image of an AuNR dimer with 2 nm gap.

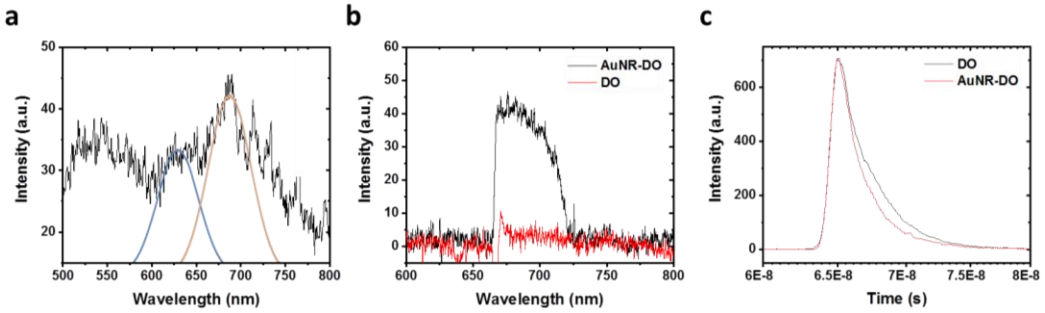

Figure 3: Optical analysis from the emitter in the nanocavity. (a) Dark field scattering spectrum that showing splitting peaks from the emitter and AuNRs. (b) An overlay of the fluorescent emission of a Cy5 dye in the nanocavity and on DO only. (c) Lifetime measurement of the Cy5 dye on DO and in the nanocavity.