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DNA-Templated Optical Nanoantennas for Single-Molecule Emission Modulation

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Optical antennas serve to enhance the interaction between light and matter by focusing freely propagating light into localized environments or facilitating the transfer of energy from localized emitters to free space. By combining with optical antennas, certain limitations associated with single emitters, such as wide emission solid angles caused by dipolar behavior and broad emission spectra resulting from multiple vibronic levels, can be overcome. However, the majority of research in these fields has focused on ensembles or diluted bulk solutions of emitters or utilizing antennas with dimensions beyond the wavelength of light. The advent of DNA origami, a bottom-up self-assembly technique, has allowed for precise control over the placement of single emitters and the construction of antennas with nanoscale precision and stoichiometry.

To tune the angular and energy distribution of emitted photons, a T-shaped DNA origami template was utilized to fabricate gold nanorod dimer antennas and nanorod monomer antennas, respectively. The robust unidirectional emission performance of a single ATTO 594 molecule, which is attached to in close proximity to a side-by-side nanorod dimer asymmetrically, has been demonstrated through numerical simulations and back focal plane imaging. The radiation pattern of the ultracompact dimer antennas exhibits a single lobe on the back focal plane, achieving a maximum forward to backward ratio of approximately 10 dB. Meanwhile, when different-sized monomer nanorods couple with single fluorophores, the emission spectra display peaks at various spectral positions due to the wavelength-dependent enhanced radiative decay rate. The modulation strength experienced by the spectra of single emitters depends on the relative position of the emitter with respect to the nanorod. Surprisingly, the fluorescence's primary and secondary peaks can be significantly enhanced or suppressed simultaneously, indicating that existing theory may underestimate the spectral reshaping effect of antennas on single molecules.

The investigation into directional emission and spectral reshaping effects of antennas opens up new possibilities in the realm of single-photon sources and their potential applications in biosensing, optical wireless links, and quantum circuits.

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