Exploring exciton dynamics in DNA-chromophore assemblies

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Control over the spatiotemporal evolution of molecular excited states is critical for the development of excitonic systems for light-harvesting, computing, and imaging. Natural light-harvesting systems achieve the required control through precise chromophore placement; however, this has been challenging to emulate in synthetic systems. Here we use DNA-chromophore assemblies to generate delocalized excitonic circuits with nanometer-scale precision over chromophore placement and orientation. Using time-resolved spectroscopy, we show how chromophore placement within a tunable DNA scaffold can be used to control electronic coupling and system-bath coupling independently. With this platform, we explore the role of these parameters in mediating exciton transport. Collectively, systematic investigation and control over excitons and their dynamics with DNA nanostructures, as presented here, offers design principles for light-harvesting and nanophotonic devices.