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Spectroscopy and Topological Phases for Organic Excitons



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abstract

The understanding and control of energy flow at the nanoscale via exciton dynamics is of fundamental chemical and physical interest, but is also technologically relevant for the design of novel light-harvesting materials. First, I will explain some of our work designing spectroscopic protocols to understand exciton dynamics under coherent illumination via ultrafast Quantum Process Tomography (QPT), a technique which retrieves the time evolution of the quantum state of excitons via nonlinear spectroscopy (1,2). As an application, I will describe the first ultrafast QPT experiment carried out with the Nelson and Bawendi groups at MIT on a nanotubular J-aggregate system at room temperature. I will also clarify the possible relevance of strongly coupled chromophores in natural light-harvesting under incoherent illumination from sunlight (3). I will explain how one can in principle distinguish excitonic coherences and their vibrational counterparts in nonlinear spectroscopy (4,5). Then, I will describe current work (6) designing topologically nontrivial phases that robustly and selectively move excitons in particular spatial directions of a molecular crystal, simulating solid state “topologically protected” phenomena like the Quantum Hall Effect, which are robust against material imperfections and static disorder. I will end by presenting our most recent work on creating one-way waveguides of plexitons (strongly coupled excitons and surface-plasmon polaritons).