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Silicon Quantum Dot Mesomaterials for Solar Energy Harvesting

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Recent progress in understanding electronic wave functions in condensed matter nanostructures has led to an ability to synthesize isolated, quantum confined building blocks with a variety of tailored optical properties. No matter what optical gap is engineered and how cleverly exciton energy is redistributed, though, novel materials composed of such nanostructures need to also exhibit efficient carrier dynamics. Transport of energy and charge is now the central issue in harnessing the true power of quantum dot materials for solar and many other uses. This is a critical bottleneck in the science because charge and exciton transport tend to proceed via low mobility, incoherent hopping associated with weak electronic coupling and high reorganization energies in these nanostructures. A number of promising strategies seek to improve energy and charge transport between quantum dots by focusing on important properties such as translational symmetry, electronic overlap, matrix encapsulation, and crystalline orientation. Our approach, though, is to consider the entire assembly as a quantum dot mesomaterial (QDM), wherein entirely new transport physics may emerge from the complex interactions between components. For instance, the superb exciton harvesting efficiency of photosynthetic complexes is at least partly due to conditions that support an element of coherent character for exciton transport. Here proteins and pigments are exquisitely structured and combined so that they perform a number of integrated functionse.g. proteins serve to correlate electronic excitations on neighboring pigments, supporting coherence and allowing exciton transport with a degree of wave-like character. We seek to design materials composed of quantum dots in which components may carry out integrated tasks that optimize dynamics ranging from incoherent random walks to coherent transport. An emphasis is placed on the robustness of such transport in the face of geometric uncertainties intrinsic to synthesized systems. The computational facet of our investigation, emphasized in this talk, utilizes an open dissipative system approach, wherein a cumulant expansion strategy is used to approximate the quantum Liouville equation via a hierarchy of density operators. This has been successfully employed to scrutinize partially coherent transport in protein/pigment complexes, but here we focus on silicon quantum dot mesomaterials and use excited state many-body calculations to populate the associated meta-Hamiltonian. After an overview of the mesomaterial perspective, this talk will focus on our computational assessment of the prospects for partially coherent exciton transport through these silicon quantum dot mesomaterials.