Excitons in Artificial Light-Harvesting Antenna Systems — Dörthe M. Eisele\(^1\), Dylan H. Arias\(^1\), Colby P. Steiner\(^1\), Robert J. Silbey\(^1\), Xiaofeng Fu\(^2\), Daniela Nicastro\(^2\), Keith A. Nelson\(^1\), and Moungi G. Bawendi\(^1\) — \(^1\)Massachusetts Institute of Technology, Cambridge, USA — \(^2\)Brandeis University, Waltham, USA

Molecular dye aggregates are close analogs of biological light-harvesting systems (LHS), and their hybrids with colloidal semiconductor quantum dots (QDs) extend the analogy to include reaction centers (RC). By means of Cryo-TEM, linear spectroscopy, and nonlinear 2D electronic spectroscopy, we show that nanotubular dye aggregates consist of two separate, weakly coupled exciton systems. In contrast, packages of these nanotubes, formed through supplementary synthetic methods, no longer consist of separate excitonic systems but rather of one strongly coupled system. These results elucidate that excitonic interactions in such nanoscale systems are not only highly sensitive to changes within the supramolecular structure, but also to changes in their higher ordering. For inorganic/organic hybrid systems we show that light absorption by dye aggregates (to mimic a LHS) followed by emission from electrostatically conjugated QDs (to mimic a RC) provides a platform to address fundamental questions of what properties control the energy transport processes in excitonic nanoscale systems.