Femtosecond electronic relaxation in cylindrical molecular aggregates

ANDREW MORAN, JORDAN WOMICK, STEPHEN MILLER, University of North Carolina — Natural light harvesting systems have evolved with correlated pigment fluctuations whose fine-tuning promotes efficient energy transfer and photosynthesis. We investigate similar correlations in a double-walled cylindrical molecular aggregate with a diameter of 10 nm. A variety of nonlinear laser spectroscopies are utilized in this work. Excitons localized on different regions of the cylindrical structure are found to undergo correlated energy level fluctuations by analysis of photon echo line shapes. Particular electronic relaxation channels are resolved with a specialized coherent Raman spectroscopy. The importance of correlated pigment fluctuations for excitonic energy transfer is discussed.