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Exciton dynamics in organic molecular crystals and nanostructures

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The photophysical behavior of organic semiconductors is governed by their excitonic states. In this talk, we classify the three different exciton types (Frenkel singlet, Frenkel triplet, and charge-transfer) typically encountered in organic semiconductors. The availability of several different exciton bands provides the possibility of interband processes. One such process is singlet fission, where an initially excited singlet exciton can spontaneously split into a pair of spin-entangled triplet excitons. We analyze this phenomenon in detail, emphasizing the role of spin state coherence and magnetic fields in studying singlet \leftrightarrow triplet pair interconversion. Singlet fission provides an example of how all three types of excitons (triplet, singlet, and charge-transfer) interact to generate unique nonlinear excitonic processes in molecular systems. These processes may be useful for applications like solar energy conversion, where the generation of two excitons per absorbed photon could lead to significant enhancements in the efficiency of single junction photovoltaic cells. Finally, we will briefly describe how excitons can also be used to initiate photochemical reactions in molecular crystal nanostructures, resulting in large shape changes and deformations.