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One Dimensional Coherent Excitons in Crystalline Organic Semiconductors

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Exciton coherence promotes long exciton diffusion lengths, associated with enhanced carrier mobilities and observations of superradiance in semiconductors. In carbon-based systems exciton coherence lengths in the micron range are observed in disorder-free systems such as single polymer chains. We focus specifically on exploring exciton coherence in small organic molecular semiconductors such as phthalocyanine derivatives (H_2OBPc), merocyanine derivatives (HB194) and quinacridone (QA). Crystalline thin films of H_2OBPc were prepared using solution processed deposition techniques, which result in macroscopic long-range order. Spin-coated HB194 thin films exhibit macroscopic three-dimensional crystals whereas vapored deposited QA films are polycrystalline.

Spatially-, temporally- and polarization-resolved photoluminescence (PL) experiments on H_2OBPc reveal the existence of a delocalized singlet exciton polarized along the high mobility axis in this quasi-1D electronic system. Furthermore, the temperature dependent PL spectroscopy of H_2OBPc is consistent with that of one-dimensional coherent excitons in a J-like aggregate semiconductor, characterized by a coherence length of approximately 32 nm at low temperatures. The coherence length is controlled by an interplay between the nearest neighbor intermolecular interaction and the exciton-optical phonon coupling which are comparable in magnitude in this system. Confocal Raman spectroscopy confirms the presence of a ~200cm^{-1} strong optical phonon mode, most likely responsible for the destruction of the coherent state at elevated temperatures.

In contrast, excitons in HB194 are localized on dimers that exhibit large coupling between the two molecules within the dimer and negligible coupling between dimers. In QA the evolution with temperature of photoluminescence is heavily affected by disorder, and experiments cannot unambiguously resolve the presence of coherent excitons.