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Coherent photoluminescence excitation spectroscopy of semicrystalline polymeric semiconductors CARLOS SILVA, PASCAL GRÉGOIRE, FÉLIX THOUIN, Univ of Montreal — In polymeric semiconductors, the competition between through-bond (intrachain) and through-space (interchain) electronic coupling determines two-dimensional spatial coherence of excitons. The balance of intra- and interchain excitonic coupling depends very sensitively on solid-state microstructure of the polymer film (polycrystalline, semicrystalline with amorphous domains, etc.). Regioregular poly(3-hexylthiophene) has emerged as a model material because its photoluminescence (PL) spectral lineshape reveals intricate information on the magnitude of excitonic coupling, the extent of energetic disorder, and on the extent to which the disordered energy landscape is correlated. I discuss implementation of coherent two-dimensional electronic spectroscopy. We identify cross peaks between 0–0 and 0–1 excitation peaks, and we measure their time evolution, which we interpret within the context of a hybrid HJ aggregate model. By measurement of the homogeneous linewidth in diverse polymer microstructures, we address the nature of optical transitions within such hynbrid aggregate model. These depend strongly on sample processing, and I discuss the relationship between microstructure, steady-state absorption and PL spectral lineshape, and 2D coherent PL excitation spectral lineshapes.

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