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Imaging heterogeneous ultrafast exciton dynamics in organic semiconducting thin films

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In solid state semiconducting molecular materials used in electro-optical applications, relatively long exciton diffusion lengths hold the promise to boost device performance by relaxing proximity constraints on the locations for light absorption and interfacial charge separation. The architecture of such materials determines their optical and electronic properties as a result of spacing- and orientation-dependent Coulomb couplings between adjacent molecules. Exciton character and dynamics are generally inferred from bulk optical measurements, which can present a severe limitation on our understanding of these films because their constituent molecules are not perfectly ordered. Rather, films of small organic molecules are composed of multiple microcrystalline domains, and this deposition-dependent microstructure can have profound impacts on transport properties. Using ultrafast transient absorption microscopy, we track the time evolution of excitons, domain by domain, in solid state thin films of TIPS-pentacene, a small soluble molecule that has recently been used in organic semiconducting devices because of its high hole mobility. The results from this spatially-resolved nonlinear optical spectroscopy support our hypothesis that bulk optical measurements deleteriously average over heterogeneities in both spatial and electronic structure; we have revealed significant inhomogeneity in exciton dynamics. Domains that appear homogeneous in linear optical microscopy are shown to have spatial variation and defects, and notable differences in exciton character and behavior are observed at domain boundaries. To interpret the contrast we observe with ultrafast dynamics, we correlate our data to local linear absorption, polarization analysis, profilometry, and atomic force microscopy. With this combined approach, we aim to ultimately understand fundamental structure-function relationship in molecular materials to provide predictive power to material development and device efficiency.