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Uncovering location-specific 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 neither perfectly ordered nor perfectly disordered. Nevertheless, such microstructure can have profound impacts on transport properties. The ultrafast spectroscopy of single domains of polycrystalline films of TIPS-pentacene, a small-molecule organic semiconductor of interest in electronic and photovoltaic applications, is investigated using transient absorption microscopy. Individual domains are distinguished by their different polarization-dependent linear and nonlinear optical responses. As compared to bulk measurements, we show that the nonlinear response within a given domain can be tied more concretely to specific physical processes that transfer exciton populations between specified electronic states. By use of this approach and a simple kinetic model, the signatures of singlet fission as well as vibrational relaxation of the initially excited singlet state are identified. As such, observing exciton dynamics within and comparing exciton dynamics between different TIPS-pentacene domains reveal the relationship between photophysics and film morphology and the potential to resolve unique signatures at interfaces between different regions of the film.