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Measuring Exciton Migration in Conjugated Polymer Films with Ultrafast Time Resolved Stimulated Emission Depletion Microscopy

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Conjugated polymers are highly tunable organic semiconductors, which can be solution processed to form thin films, making them prime candidates for organic photovoltaic devices. One of the most important parameters in a conjugated polymer solar cell is the exciton diffusion length, which depends on intermolecular couplings, and is typically on the order of 10 nm. This mean exciton migration can vary dramatically between films and within a single film due to heterogeneities in morphology on length scales of 10's to 100's nm. To study the variability of exciton diffusion and morphology within individual conjugated polymer films, we are adapting stimulated emission depletion (STED) microscopy. STED is typically used in biology with sparse well-engineered fluorescent labels or on NV-centers in diamond. I will, however, describe how we have demonstrated the extension of STED to conjugated polymer films and nanoparticles of MEH-PPV and CN-PPV, despite the presence of two photon absorption, by taking care to first understand the material's photophysical properties. We then further adapt this approach, by introducing a second ultrafast STED pulse at a variable delay. Excitons that migrate away from the initial subdiffraction excitation volume during the ps-ns time delay, are preferentially quenched by the second STED pulse, while those that remain in the initial volume survive. The resulting effect of the second STED pulse is modulated by the degree of migration over the ultrafast time delay, thus providing a new method to study exciton migration. Since this technique utilizes subdiffraction optical excitation and detection volumes with ultrafast time resolution, it provides a means of spatially and temporally resolving measurements of exciton migration on the native length and time scales. In this way, we will obtain a spatiotemporal map of exciton distributions and migration that will help to correlate the energetic landscape to film morphology at the nanoscale.