

Added Value of Circular Dichroism for Studying Charge Transfer Excitons in Photoactive Organic Thin Films

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Organic semiconductors have emerged as promising materials for various optoelectronic applications due to their unique properties and easy adjustability. For such optoelectronic devices, however, it is essential to acquire a fundamental understanding of the underlying processes and mechanisms such as the generation, transport and nature of excitons. *N*-alkyl anilino squaraines are prototypical materials for such applications, featuring distinctive double humped absorption spectra.^[1] To explain this absorption pattern by theoretical modeling, two different approaches have been used so far. The first theoretical model is an essential state model which uses intermolecular Frenkel excitons in combination with intermolecular charge transfer excitons.^[2,3] The other suitable model, however, assumes an ensemble of aggregates with intermolecular dipole–dipole resonance interactions and structural disorder in the thin film to describe the unpolarized absorbance spectrum.^[4]

Here, we investigate the structure-correlated excitonic properties in photoactive thin-film semiconductors for optoelectronics by comparing non-chiral nBSQ and nOSQ to two new structurally related chiral anilino squaraines. Substitution of only one *n*-alkyl side chain with an enantiomerically pure citronellyl-derived residue ensures the analogy to the corresponding *n*-alkylated compounds. The presence of the same double hump absorption features allows comparison with the *n*-alkyl derivatives, while an emerging excitonic chiral dichroism (CD) simultaneously enables investigation by Mueller matrix polarimetry. These dyes are thus predestined to provide a deeper insight into the nature of excitonic transitions between aggregated squaraine molecules.

References:

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