

Single-object polarization-resolved microscopy on artificial light-harvesting complexes

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Natural light-harvesting antennae, found throughout photosynthetic organisms such as green algae and bacteria, achieve remarkable energy-transfer efficiencies through the dense packing and strong mutual coupling of chromophores. This organization gives rise to delocalized Frenkel excitons, whose migration through the antenna complex enables highly efficient excitation-energy transport [1]. Molecular double-walled nanotubes (DWNTs), formed by the self-assembly of amphiphilic C8S3 dye molecules in aqueous solution, offer a structurally well-defined biomimetic platform for studying these processes. Although they are soft molecular assemblies, DWNTs display an unusually low level of structural disorder, permitting exciton delocalization over hundreds of chromophores. Realizing the full potential of such systems requires experimental approaches that provide spectroscopic observables directly comparable with theory and, at the same time, establish how robust exciton delocalization remains under structural or energetic perturbations.

In this contribution, I discuss selected examples of polarization-resolved photoluminescence microscopy on individual DWNTs. I first show that the transition dipole moments of the emitting excitons are confined to a narrow angular distribution oriented along the nanotube axis. This effect originates from negative intermolecular couplings, which favor exciton states with in-phase wavefunction coefficients and yield a reduced linear dichroism close to unity [2]. I then show that Frenkel excitons in DWNTs retain extensive delocalization even when substitutional molecular dopants, acting as deep energetic traps, are incorporated into the assembly. Importantly, this behavior is not limited to photoluminescence but is also observed in absorption, demonstrating that it reflects the intrinsic excitonic structure rather than a selective subset of emissive states. Theoretical simulations capture the observed behavior and indicate that excitons redistribute adaptively to avoid trap sites [3]. Together, these results demonstrate the resilience of supramolecular nanotube architectures in sustaining efficient exciton transport under conditions relevant to molecular fabrication.

[1] Jansen *et al.*, *J. Chem. Phys. Rev.* 5, 041305 (2024)

[2] Kuevda *et al.*, *JACS* 147, 6171 (2025)

[3] Kuevda *et al.*, <https://chemrxiv.org/doi/full/10.26434/chemrxiv-2025-p4jrf/v4> (2026)

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