Title:	Unravelling the Role of Water in Ultrafast Excited-State Relaxation Dynamics within Nano-Architectures of Chlorophyll a
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Abstract:	Water plays a pivotal role in structural stability of supramolecular pigment assemblies designed for natural light harvesting (for example, chlorosome antenna complex) as well as their artificial analogs. However, the dynamic role of water in the context of excite-state relaxation has not been explored till date, which we report here. Using femtosecond transient absorption spectroscopy, we investigate the excited-state dynamics of two types of nano-scale assemblies of chlorophyll a with different structural motifs, rod-shaped and micellar assemblies, that depend on the water content. We show how water participates in excess energy dissipation by vibrational cooling of the non-thermally populated Qy band at different rates in different types of clusters but exhibits no polar solvation dynamics. For the micelles, we observe a bifurcation of stimulated emission line shape, whereas a positive-to-negative switching of differential absorption is observed for the rods; both these observations are correlated with their specific structural aspects. Density functional theory calculations reveal two possible stable ground state geometries of dimers, accounting for the bifurcation of line shape in micelles. Thus, our study elucidates water-mediated structure–function relationship within these pigment assemblies.
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