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Title:	Tuning effect of local environment to control mechanism of fluorescence depolarization: Rotational diffusion and resonance energy transfer within homo-aggregates of xanthenes
Authors:	Silori, Y. (/jspui/browse?type=author&value=Silori%2C+Y.) De, A.K. (/jspui/browse?type=author&value=De%2C+A.K.)
Keywords:	Molecular aggregates Time-resolved fluorescence depolarization Rotational diffusion Förster resonance energy transfer (FRET)
Issue Date:	2019
Publisher:	Elsevier
Citation:	Journal of Photochemistry and Photobiology A: Chemistry, 377, pp. 198-206.
Abstract:	Controlling excitation energy transfer within self-assembled molecular aggregates has been a major challenge since such systems represent as prototypes for highly efficient light-harvesting pigment assemblies within photosynthetic organisms. Using time-resolved fluorescence depolarization studies, here we report complete reversal of trends in anisotropy decay traces up on increasing fluorophore concentration while traversing a series of solvent mixtures (water and glycerol) with increasing viscosity. For glycerol, in dilute solution depolarization results from rotational diffusion whereas Förster resonance energy transfer (FRET) is found to be the key mechanism in concentrated solution; in contrast, for water, rotational diffusion is the dominant reason for fluorescence depolarization at all concentrations. We further investigate how viscosity, rather than dielectric constant of the solvent, controls contribution from FRET towards depolarization by partially immobilizing the fluorophores. The results are explained within the framework of analytical models for fluorescence depolarization due to either rotational diffusion or FRET. We further examine the unusual 'dip-and-rise' behavior in anisotropy traces for intermediate range of concentration which is suggestive of presence of heterogeneity within homochromophoric system. These results show how one can fine-tune the explicit nature of solvent to facilitate FRET over other competing dynamical processes.
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