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Title: Ultrafast Excited-State Dynamics of Tricarbocyanine Dyes Probed by Two-Dimensional Electronic

Spectroscopy: Polar Solvation vs Photoisomerization

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Abstract:

Photophysical properties of tricarbocyanine dyes in various solvents have been widely investigated using a variety of spectroscopic tools. However, the presence of several ground-state isomers and interconversion between these isomers on an ultrafast timescale upon photoexcitation render unambiguous assignment of spectral features guite difficult. In this work, ultrafast excited-state dynamics of two tricarbocyanine dyes in two solvents, DNTTCI and IR140, in ethanol and ethylene glycol, are studied by two-dimensional electronic spectroscopy (2DES). We present a detailed discussion on design and calibration of the 2DES apparatus and on the method for data processing by phase-cycling. For DNTTCI we report a method to obtain solvation correlation function, the nature of which is found to be strongly dependent on the excitation frequencies; a blue-shifted spectrum at early time is observed and explained based on preferential emission from a subset among various isomers having overlapping spectral features. For IR140 in ethanol, four isomers with distinct spectral features are identified, and most importantly, three of these isomers were found to interconvert upon photoexcitation which completes within 100 fs and is explained based on a kinetic model of consecutive chemical reaction. Density functional theory calculations show the presence of several ground-state isomers for both these dyes. Through this work we demonstrate how 2DES can help us to decipher distinct excited-state photophysics in two carbocyanine dyes, polar solvation and photoisomerization, by resolving spectral congestion without sacrificing time resolution.

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