Ultrafast coherent vibronic dynamics in organic conjugated molecules

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Photoinduced processes of organic conjugated materials frequently involve concerted dynamics of coupled electronic and vibrational degrees of freedom (i.e. vibronic couplings) that can give rise to persisting phase relations or coherences. Fundamental insights into the coherence creation and destruction mechanisms can potentially allow the manipulation of photoexcited non-radiative pathways to achieve desired efficient transfer of energy and charges.

Direct quantum molecular simulations using the ab initio multiple cloning (AIMC) approach [1,2] is a controllable approximation to non-adiabatic dynamics and naturally includes electronic decoherence. Their modeling results can be used to predict nonlinear X-ray signals that track its coherent behavior.

Herein, AIMC have been applied to analyze coherences during the inter-chromophore energy transfer of two different organic conjugated materials: (a) a rigid synthetic molecular heterodimer presenting persistent coherence[3,4]; (b) a flexible dendrimer whose initial ultrafast coherent dynamics is followed by incoherent mechanisms governed by thermal fluctuations that ultimately lead to a random molecular scrambling[5].

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