MAR14-2014-021058

Abstract for an Invited Paper
for the MAR14 Meeting of
the American Physical Society

Mapping Energy Flow with Ultrafast Optical Spectroscopy

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Ultrafast electronic spectroscopy connects the spatial, temporal and dynamic landscapes of complex systems. These connections are essential to our understanding of structure-function relationships and energy transport through materials. Using two-dimensional electronic spectroscopy (2DES), we can generate correlation maps that relate the initial absorptive interaction with the signal emission, allowing us to follow the flow of energy through a system via the vibrational and electronic coherences and populations. Using 2DES to study synthetic and natural photosynthetic pigments provides insight into the advantages of particular molecular architectures that are ubiquitous in nature, optimizing efficient energy transfer and demonstrating the importance of static disorder and vibrational coupling. The critical role of vibrations in these pigments is mirrored in the response of the nitrogen vacancy centers in diamond (NV-diamond) quantum material system. 2DES studies of NV-diamond reveal an array of coherent nuclear vibrations coupled to the electronic state. The effect of the vibrational coherences on the dynamics of the NV-diamond system may provide a route to increased efficiency of energy transport in nanostructured solar cells.