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Title:	Probing Intramolecular Energy and Charge Transfer using Broadband Pump-Probe Spectroscopy and Impulsive Stimulated Raman Spectroscopy
Authors:	Dhamija, Shaina
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Abstract:	<p>Abstract Photoinduced processes involving energy and charge transfer, for example, photosynthetic light-harvesting and photoisomerization in receptor protein rhodopsin, are indispensable for driving life processes. This necessitates the study of model systems in order to provide a molecular-level understanding of such processes. Ultrafast spectroscopy, using a tailored sequence of femtosecond laser pulses at optical frequencies, has been used to probe the relaxation dynamics of the photo-initiated electronic excited state which is often accompanied by structural changes. Real-time monitoring of such changes in molecular geometry has been a long sought-after goal. However, even for small chromophores in solution, identifying the origin of these vibrations, i.e., whether they arise from ground or excited electronic state of the solute, or contributed by ground electronic state of the solvent as well, has been debatable. This thesis focuses on exploring photoinduced intramolecular energy and charge transfer and providing a systematic methodology to pinpoint the origin of vibrations, using third-order non-linear spectroscopic techniques, i.e. broadband pump-probe spectroscopy and impulsive stimulated Raman spectroscopy. Following an explanation of this project's impetus and complete description of the instrumentation procedures, excitonic coupling in bis-BODIPY complexes of hexaphyrin-like macrocycles will be presented. In the next part, photoinduced phenomena in fluorescent proteins involving light-driven proton transfer and/or electron transfer and structurally flexible stilbene derivatives undergoing electron transfer will be discussed. Further, to identify the key vibrations leading to structural changes, a general methodology developed to disentangle the origin of vibrations for diatomic and polyatomic molecules in solution will be discussed. At last, a discussion on future prospects is provided.</p>
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