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Femtosecond Stimulated Raman Spectroscopy by Six-Wave Mixing

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Knowledge of the structural changes that accompany photochemical reactions has motivated the development of a wide variety of time-resolved vibrational spectroscopies. For example, a technique known as femtosecond stimulated Raman spectroscopy (FSRS) has yielded important insights into numerous photochemical processes in the past 10-15 years. Simultaneous probing of all resonances in the fingerprint region of the vibrational spectrum and sensitivity to dynamics on the 100-fs time scale are the primary selling points for the FSRS technique. Despite its utility, FSRS is challenged by a large background of residual laser light and lower-order nonlinearities. In this talk, I will introduce a newly developed FSRS experiment in which five laser beams are used to eliminate the background of residual laser light and lower-order nonlinearities present in the traditional three-beam FSRS geometry. Applications to photodissociation reactions in triiodide and heme proteins will be discussed. It is envisioned that this approach will be useful for investigating photoinduced dynamics in a wide variety of condensed phase systems.