## **Two-Dimensional Raman Spectroscopy of Metallocenes:** Probing the Dynamics of Organometallic Bonds

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Vibrational coupling in aromatic hydrocarbons and metallocenes will be probed using femtosecond stimulated Raman spectroscopy (FSRS). FSRS has a unique ability to introduce and probe vibrational coherence in order to elucidate the anharmonic vibrational couplings that drive energy flow through molecules.





(a) Schematic of 2D-FSRS experiment. (b) The detected spectrum of the probe measures the interaction between the impulsively driven vibrations and the vibrations probed by FSRS. In the presence of anharmonic coupling between the driven ( $\omega_{low}$ ) and probed ( $\omega_{hi}$ ) vibrations or via a coherent cascade, sidebands are observed in the stimulated Raman spectrum at Raman shifts of  $\omega = \omega^{(+)} = \omega_{hi} + \omega_{low}$  and  $\omega = \omega^{(-)} = \omega_{hi} - \omega_{low}$ . The shape of the sideband peak varies with changes in time delay,  $\Delta t$ , and produces a time dependent lineshape.