Femtosecond Frequency Domain Multiphoton Pump-CMDS Probe Spectroscopy of Transition Metal Dichalcogenide, Perovskite, and Nanoscale Heterostructures, and Enzymatic Molecular Systems

John C. Wright, Department of Chemistry, University of Wisconsin, Madison, WI

wright@chem.wisc.edu

Abstract

The talk describes pump-fully coherent CMDS probe spectroscopy where a 1-2 photon pump creates a population and a 3-photon probe creates a fully coherent Schrodinger cat state containing triple quantum vibrational and/or electronic coherences. The cat state collapse insures the 3-photon probe is an instantaneous snap-shot that isolates the coupled quantum states population dynamics within the pump-probe delay time. We applied the methodology to follow the femtosecond charge dynamics of $MoSe_2$, WS_2, 2D and 3D perovskites, and their heterostructures. We demonstrate how we extract the density of states within the Brillouin zone. Similarly, application to enzymatic molecular systems identifies the coupled vibrational reaction coordinates on the electronic potential energy surface.